Electron-hole liquid in germanium under infinite uniaxial compression*

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We present analysis of a series of luminescence spectra arising from the electron-hole liquid in germanium under uniaxial compressive stress oriented along the [111] crystallographic axis. We evaluate the ground-state properties of the liquid in the infinite-stress limit using an extrapolation. We find that the electron Fermi energy is reduced from its zero-stress value by a factor of 3.3, the hole Fermi energy by 2.0, and the e-h pair density by 24, in agreement with theoretical predictions.

I. INTRODUCTION

A uniaxial compressive stress applied to germanium along the [111] crystallographic axis leads to a considerable simplification of the band structure in the large-stress limit. The degeneracy among the four conduction electron valleys is lifted as the valley aligned along the stress axis is lowered in energy while the other valleys are raised. Likewise, the valence band is split into two branches with the lower-mass band shifted to higher energy so that it is preferentially occupied by holes. However, the hole bands retain substantial anisotropy and energy dependence to their effective mass, until stresses greater than about 100 kg/mm^2 are attained.

Early studies of the electron-hole (e-h) liquid by Bagaev, Galkina, Gogolin, and Keldysh,[1] by Benoît à la Guillaume, Voos, and Salvan,[2] and by others[3,4] have shown that the condensed phase is modified by stress as evidenced by a shift in the peak position and a change in linewidth and intensity of the luminescence. These workers pointed out that the observed changes were qualitatively consistent with a decrease in the e-h liquid binding energy with respect to excitons and with a decrease in the liquid density, both of which would be expected from the decrease in the electron and hole density of states with stress.

Recently Feldman, Chou, and Wong[5] have analyzed the e-h liquid (and exciton) luminescence more carefully and obtained reliable values of the ground-state energies and density at a single value of stress, with a closer approach to stress homogeneity than in some of the previous work. We have performed similar quantitative analyses at a series of values of uniaxial [111] stress and extrapolated to the infinite stress limit. We emphasize, as have Markiewicz and Kelso,[6] that such an extrapolation is necessary because the highest stresses used in existing data are too small by about an order of magnitude to approximate closely the infinite stress limit for the e-h liquid.

In Sec. II we describe the experimental procedures with emphasis on the method used to approach stress homogeneity and to test for such a condition. In Sec. III we discuss the data analysis and indicate some useful approximations for the hole bands. Finally, in Sec. IV, we compare the results with theory and with published experiments.

II. EXPERIMENTAL CONDITIONS

The most important aspect of uniaxial stress measurements is the homogeneity of deformation. We have attempted to maximize this uniformity by using the center of a long thin sample with pointed ends and have tested for success of the procedure by monitoring the e-h liquid luminescence intensity as a function of stress.

The stress chamber is illustrated schematically in Fig. 1. The samples used were cut from a larger boule to a size 15 mm long and 2.5×1.7 mm in cross section with their long axis carefully aligned along the [111] crystallographic axis and the faces polished flat and parallel. The crystals were illuminated near their center with 0.5145-μm-wavelength light from an argon laser. The stress was applied using a balance beam at the top of the cryostat with calibrated weights hung from the long arm. Further experimental details are discussed by Pokrovskii and Svistunova.[4]

A series of samples was tested for stress homogeneity by monitoring the e-h liquid luminescence intensity variation and a sample was selected for which the intensity increased with stress in a way similar to that described by Pokrovskii and Svistunova.[7] We would argue that this intensity increase suggests that the results here are

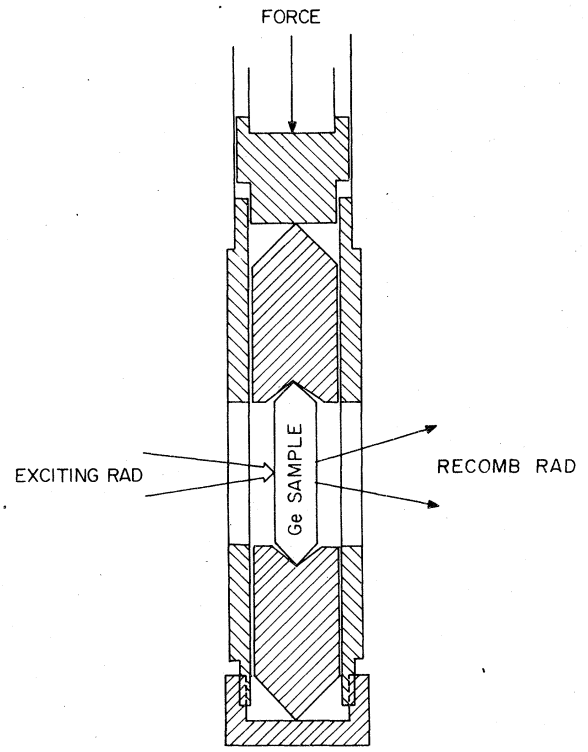

FIG. 1. Schematic drawing of the apparatus for application of stress showing the pointed piston and sample arrangement used to enhance the stress homogeneity near the center of the 15×2.5×1.7 mm crystal oriented along its [111] axis.

representative of more-nearly-uniform stress S than other studies in which the intensity was found to decrease. (An exception to this criterion is that substantial strain gradients can still be present in the case where a strain pattern confines the liquid in a region away from the surfaces near the point of excitation.[6]) We expect that an intensity increase should occur as the e-h liquid density decreases, at temperatures well below the critical point, because of an increase in the e-h liquid lifetime and radiative efficiency. On the other hand, nonuniform stress can reduce the intensity by either moving the e-h drops to the sample surface where they decay nonradiatively or by heating the liquid by motion without interaction with the surface. In the latter case, the electron-lattice interaction, which has a damping component, can lead to substantial evaporation of the liquid, particularly if droplet velocities approach the speed of sound.[3,7]

Other aspects of our data seem to confirm the relative stress uniformity in our samples. First, with stress the exciton luminescence line does not show any additional broadening in measurements at 4.2 K and the liquid line narrows in its full width at half maximum continuously in the data presented below. Second, we see no variation in the e-h liquid line with the position of the exciting laser spot or the point of the optical collection cone. Third, we observe only one (shifted) e-h liquid line associated with LA phonon emission and not a second line, unshifted in photon energy, that might arise from a region of the sample at low relative stress. In the case of continuously inhomogeneous stress, only line broadening will occur.

III. ANALYSIS OF DATA

We have analyzed the luminescence line shapes using the simple joint density of states of electrons and holes.[8] At a given value of S (along the [111] axis in all cases studied here) we have utilized reasonable approximations to the exact electron and hole bands to describe the density of states. Six values of S were studied with $0 \leq S \leq 9.50$ kg/mm^2 and the highest two and lowest two were analyzed in some detail, while only an approximate treatment was made of the intermediate cases. The four detailed cases are shown in Figs. 2–5. At all these values of S the line shapes were the same within experimental uncertainty for polarization of the emitted light parallel or perpendicular to the direction of S. In Figs. 2 and 3 ($S = 0$ and 1.25 kg/mm^2) the luminescence intensity is independent of polarization ($I_\parallel = I_\perp$) but at larger S the parallel component becomes larger, increasing to a ratio $I_\parallel/I_\perp = 1.33$ at $S = 9.50$ kg/mm^2.

We have analyzed here only the luminescence replica involving the emission of a LA phonon. The reason for our choice is that, as discussed by Martin[9] and by Thomas and Capizzi,[10] all of the other phonon replicas involve some forbidden recombination processes. The TO replica, for example, is an interesting, but complicated, case as it is composed of both forbidden processes, occuring via the Γ_2' intermediate state, and allowed processes, via the Γ_{15} and Γ_3' intermediate states. As discussed previously by Bir and Picus[11] and by Pokrovskii and Svistunova,[4] the particular combination of recombination routes determines the relative polarization intensities.

Kane[12] has calculated that, if the LA line were produced only by the Γ_2' route with no matrix-element effects, the wave-function symmetries would give $(I_\parallel/I_\perp)_{LA} = 2$ for the band-extremum recombination in the e-h liquid. Under the same assumption, the allowed part of the TO replica would give $(I_\parallel/I_\perp)_{TO} = \frac{1}{4}$ for Γ_{15} and $I_\parallel = 0$ for L_3'. In our highest stress, $S = 9.5$ kg/mm^2, we find $(I_\parallel/I_\perp)_{TO} = 0.70$. The qualitative nature of the dominant polarization for both TO and LA is given correctly by these wave-function considerations,

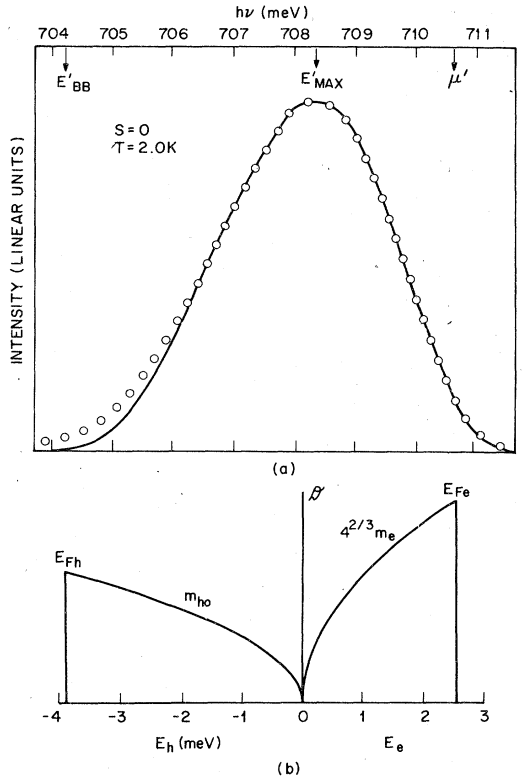

FIG. 2. Data and analysis of the LA-phonon luminescence replica from the e-h liquid in Ge at $T = 2.0$ K and stress $S = 0$. In (a), the open circles are data and the solid line is a theoretical fit using a convolution of the density of states of electrons and holes shown to scale in (b). The characteristic parameters obtained from the fit are plotted in Figs. 6 and 7 and are in agreement with previous measurements.

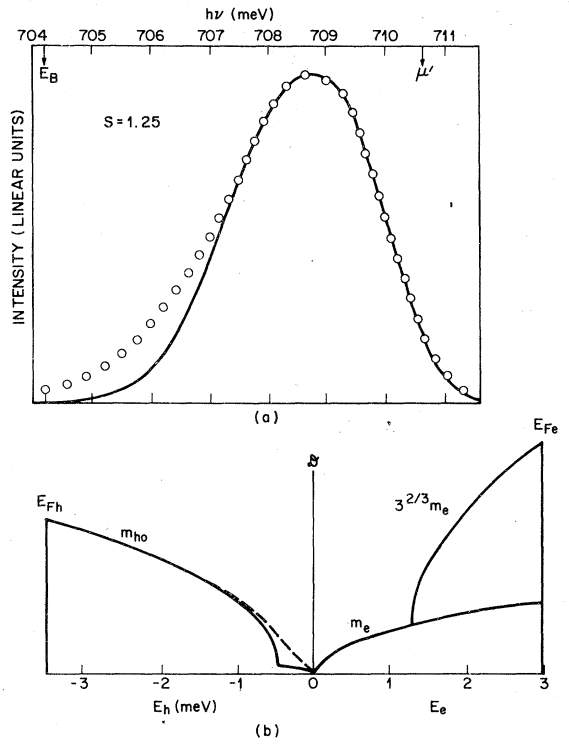

FIG. 3. Data and analysis similar to Fig. 2 at a uniaxial compressive stress $S = 1.25$ kg/mm^2 applied parallel to the [111] crystallographic axis. A rough approximation (solid curve) for the exact hole density of states (dashed curve) is used neglecting the nonparabolicity of the hole masses.

as noted previously.[4,11] The less extreme magnitudes of the polarization ratio in the data than in the model may be due to matrix element effects and, in the case of the TO line, to substantial contributions from the forbidden Γ_2' rate.

In Fig. 2 we show the analysis for the LA line at $S = 0$. In Fig. 2(a) the dots are data at $T = 2.0$ K and the solid line is the calculated fit using the density of states as shown in (b). This fit reproduces the previous results of a number of workers[2,9,13,14] and uses precisely the same parameters as those of Thomas and Capizzi.[10] The electron density of states mass,[16] $m_e = 0.22m$, where m is the free-electron mass, is used for each of the four valleys. The hole density of states mass[16] $m_{ho} = 0.357m$ is used, based on an average over the anisotropic hole bands appropriate for zero stress. The fit gives the electron Fermi energy $E_{Fe} = 2.53 \pm 0.01$ meV, hole Fermi energy $E_{Fh} = 3.90 \pm 0.01$ meV, and the e-h pair density $n = 2.39 \times 10^{17}$ cm^{-3}. Throughout our analysis, we have calculated the density using E_{Fe} and the excellent approximation

$$n(T) = 1.48 \times 10^{16} (k_B T)^{1.5} \times [(E_{Fe}/k_B T)^2 + 1.7]^{3/4} \text{ cm}^{-3},$$

with $k_B T$ in meV, for each of the four electron valleys. For upwardly shifted valleys (see below) the appropriate value of E_{Fe}, smaller than for the lower valley, is used. This formula can be obtained from an approximate solution to the usual integral[15] over the density of occupied states. We use $n(2\text{ K}) \approx n(0) \equiv n$ for comparison with the $T = 0$ theoretical results, neglecting the temperature correction which is small compared to the strain effects. The e and h Fermi energies are shown to scale in Fig. 2(b) and plotted in Fig. 6 with their sum in Fig. 7 (right-hand axis). The density n is plotted in Fig. 7 (left-hand axis).

Three characteristic photon energies are also determined by the fitting in Fig. 2: the bottom of the band $E'_{BB} = 704.2 \pm 0.5$ meV, the intensity maximum of the band $E'_{max} = 708.2 \pm 0.5$ meV, and the chemical potential $\mu' = 710.6 \pm 0.5$ meV. These values are slightly lower than those of Thomas

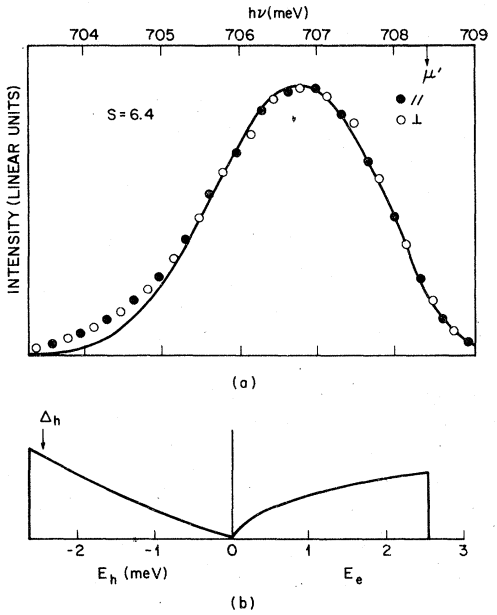

FIG. 4. Data and analysis similar to Fig. 2, but at $S = 6.40$ kg/mm^2. At this stress there is substantial polarization of the luminescence with the solid circles indicating orientation parallel to the stress axis and open circles perpendicular. The two data sets have been normalized at the line peak. A close approximation to the exact hole density of states is used as shown in (b) and discussed in the text. The lower hole band is nearly depopulated as shown by the splitting Δ_h.

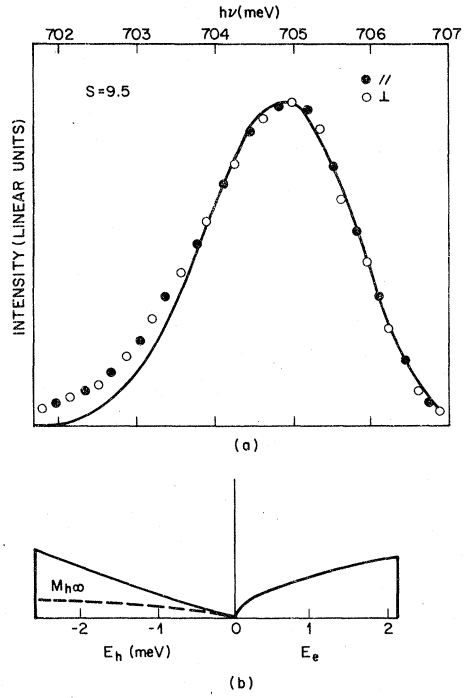

FIG. 5. Data and analysis similar to Fig. 4, but at $S = 9.5$ kg/mm^2, where there is no difference between the line shapes for parallel and perpendicular polarizations within our uncertainty. Both the holes and electrons occupy single bands at this stress but the holes' density of states still differs markedly from a $\sqrt{E}$ form. The close approximation to the exact hole bands used in the fitting is shown as the solid line in (b), where, for comparison, the infinite-stress hole density of states is shown as the dashed line.

et al.,[15] for example, but in agreement within the uncertainty limits. Our primary interest, in comparing our results with theoretical calculations, is in relative values of energy, rather than in the absolute energy positions, which we distinguish with primes.

In all of our analysis we shall not consider two refinements which have been made to the fitting. First, as discussed by Thomas and Capizzi,[10] the unresolved LO luminescence replica contributes about 8% intensity peaked near the lower edge of the LA line. If the LO line is included, $E_{Fe}+E_{Fh}$ and n are reduced by 3% and 4%. Second, as discussed by Martin and Störmer[17] and by others,[18] without consideration of the LO replica, the extra luminescence intensity on the low-energy edge of the line should be accounted for in a definitive fit. The reduction in the experimental value of n that will result from the inclusion of this extra broadening will be comparable to the other correction.

In Fig. 3(a), we show the comparison of theory and experiment for $S = 1.25$ kg/mm^2 and in Fig. 3(b) the approximation used for the density of states. The electrons have available one valley lowered by a splitting Δ_e below the other three valleys. All four valleys are partially occupied up to E_{Fe}. The

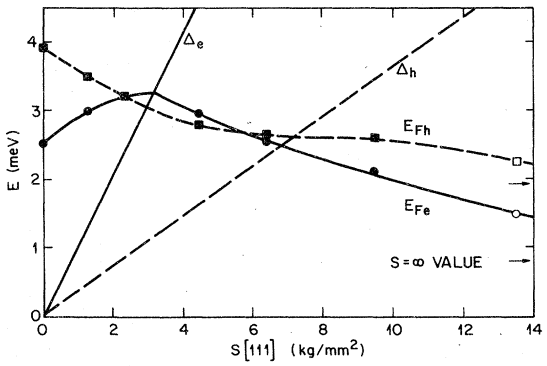

FIG. 6. Summary of the electron and hole Fermi energies E_{Fe} and E_{Fh} obtained as illustrated in Figs. 2–5. The electron and hole band splittings Δ_e and Δ_h are also shown for comparison. The dashed and solid lines through the data are visual guides only and the infinite stress values for E_{Fe} and E_{Fh}, obtained from Fig. 8 as described in the text, are indicated by arrows. The solid data points are from this work and the open points are from Ref. 5.

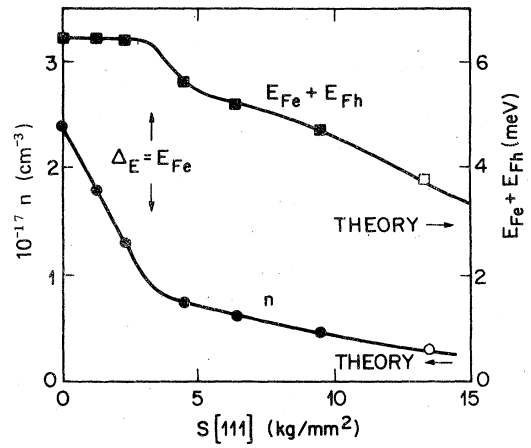

FIG. 7. Summary of results for $E_{Fe}+E_{Fh}$, right-hand axis, and n, left-hand axis, as labeled. Theoretical results of the self-consistent approximation of Ref. 22 are indicated by arrows, and are found to be in agreement with the $S=\infty$ values obtained from Fig. 8. Open data points are from Ref. 5.

value of Δ_e is determined from the relation $\Delta_e(\text{meV}) = (\frac{4}{9}S_{44}\Xi_u)S = 1.03S$ (kg/mm^2), based on the results of measurements on bulk Ge.[16,19] All valleys are assumed to retain the same effective mass as for $S=0$ and their densities of states are summed.

Although the exact density of states is more complicated for the holes than for the electrons, we have used a somewhat similar approximation, based on a suggestion by Kane.[12] We assume that there is a band with a mass change at an energy

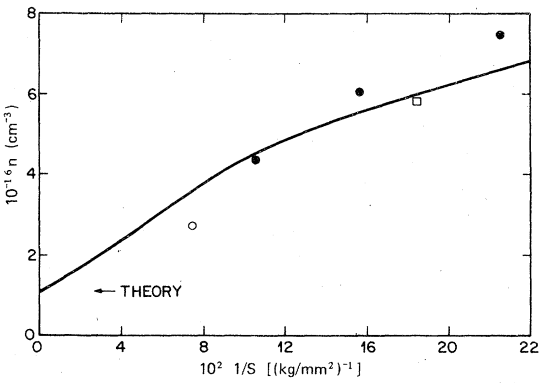

FIG. 8. Electron-hole liquid density as a function of inverse stress including only the data for $S>4.45$ kg/mm^2. The solid curve is a theoretical estimate of the expected density variation scaled in magnitude to give the fit (weighted toward higher S) to the data. The intercept of this curve at $1/S=0$ gives our extrapolated value for the density in the infinite stress limit. The circle is data from Ref. 5, the open square is theory from Ref. 25, and the arrow is theory for $1/S=0$ from Ref. 22.

Δ_h, determined in the same way as Δ_e, with the value[16] $\Delta_h(\text{meV}) = \frac{2}{3}S_{44}D_u'S = 0.364S$ (kg/mm^2). The large mass is assigned the zero-stress hole mass m_{h0} and the small mass the value determined near the band edge in large stress $m_{h\infty}=0.088m$. An inaccuracy in this approximation arises because the hole masses in both bands are energy dependent. We have illustrated the accuracy of this approximation by plotting the exact hole density of states, for comparison, as the dashed line in Fig. 3(b). The values used in Fig. 3 are $E_{Fe}=2.98$ meV, $E_{Fh}=3.49$ meV, and $n=1.80\times10^{17}$ cm^{-3}.

Less detailed analyses were carried out at $S=2.30$ and 4.45 kg/mm^2 using an interpolation between lower and higher stresses and the measured linewidths. In these two values of S the line shape differed measurably for $I_\parallel$ and $I_\perp$ so that the polarization dependence of the density of states must be considered to obtain a definitive analysis. We arbitrarily considered only $I_\parallel$ to obtain the values plotted in Figs. 6–8 with the errors assumed to be the same magnitude as for $S=1.25$ kg/mm^2. An interesting qualitative feature of the results is that E_{Fe} increases as the upper electron bands empty out up to $S=3.2\pm0.3$ kg/mm^2 where $\Delta_e=E_{Fe}=3.3$ meV.

In Fig. 4 we show the results of our analysis of the data at $S=6.4$ kg/mm^2. The solid circles are for $I_\parallel$, the open circles for $I_\perp$. In this case the simple two-hole band approximation gives an unsatisfactory fit. We have found, however, that the holes' density of states mass can be approximated quite accurately by the formula

$$m_{dh}=0.4055(E/S)^{0.45}, \quad \text{for } 0.06<E/S<0.4,$$

where E is the energy in meV and S is in kg/mm^2. The upper limit of this range of applicability just fits the energy span filled by the e-h liquid at this stress. We have plotted the exact density of states for comparison in Fig. 4(b), but the difference is not visible within the width of the solid line, even though the lower hole band is still populated. We have used the exact hole bands to determine the ratio of the electron to hole Fermi energies for the fit, since a slightly different mass, integrated over E_h, is required. The parameters shown in Fig. 4 are $E_{Fe}=2.55$ meV, $E_{Fh}=2.62$ meV, and $n=6.03\times10^{16}$ cm^{-3}.

For the data in Fig. 5, at $S=9.5$ kg/mm^2, we find that Δ_h is greater than E_{Fh} so that we have reached the single-band limit, but the holes remain strikingly nonparabolic. Again we find the approximation for m_{dh} given above to be excellent and obtain a good fit to the data as shown in Fig. 5(a). The fit yields the values $E_{Fe}=2.10$ meV, $E_{Fh}=2.58$ meV, and $n=4.53\times10^{16}$ cm^{-3}. We have

shown, for comparison, in Fig. 5(b), the infinite-stress hole density of states, and we see that we are far from this limit of parabolic hole bands.

We have also used the above approximations to reanalyze the results of Feldman, Chou, and Wong[5] at $S = 13.5$ kg/mm^2 and find $E_{Fe} = 1.49$ meV, $E_{Fh} = 2.24$ meV, and $n = 2.74 \times 10^{16}$ cm^{-3}, in agreement with their findings. We include their result along with ours in Figs. 6–8. A comparison of these independent results suggests that they are consistent with each other and with the claim that both sets of data are essentially free from the effects of strain inhomogeneities.

The trend of E_{Fh} between $S = 6.40$ and 9.50 kg/mm^2 indicates that $\Delta_h = E_{Fh} = 2.6$ meV at a point $S = 6.9$ kg/mm^2 as shown in Fig. 6. Furthermore, it is clear that neither E_{Fe} nor E_{Fh} nor n has reached its limiting value.

IV. DISCUSSION OF RESULTS

Following the initial calculation of the ground-state parameters of the e-h liquid in the infinite-stress limit by Combescot and Nozières[20] and by Brinkman and Rice,[21] Vashishta, Bhattacharyya, and Singwi[22] have performed more-detailed calculations. In the latter work, several models were used including the Hubbard approximation,[23] which gives results similar to the earlier theories, and a fully self-consistent approximation, based on the analysis of the general electron gas by Singwi, Tosi, Land, and Sjolander.[24] In the latter calculation[22] the results are $E_{Fe} = 0.8$ meV, $E_{Fh} = 2.1$ meV, and $n = 1.1 \times 10^{16}$ cm^{-3} as indicated in Figs. 6–8. The results of the Hartree approximation are similar with $E_{Fe} + E_{Fh} = 3.1$ meV and $n = 1.1 \times 10^{16}$ cm^{-3}. Although the experimental values at the highest measured stresses are larger than theory in all cases, it is clear that substantial reductions will occur at higher stress.

Liu[25] has calculated the e-h liquid density at intermediate stress $S = 5.5$ kg/mm^2, where the hole splitting $\Delta_h = 2$ meV, and finds $n = 5.8 \times 10^{16}$ cm^{-3}. We have plotted his result on Fig. 8 along with the data, and we see reasonable agreement with our results at comparable strain.

In Fig. 8 we show our approximate extrapolation to the infinite-stress limit. We have plotted n as a function of $1/S$ for the data at $S \geq 4.45$ kg/mm^2. We have used a theoretical calculation of $n(S)$, scaled in magnitude to give a fit to the four data points, as shown by the solid curve. The theoretical curve is calculated, following Markiewicz and Kelso,[6] including only the stress dependence of the kinetic energy and using an approximate Wigner-type form for the correlation energy with parameters chosen to fit theory[22] in zero stress. The intercept of this curve at $1/S = 0$ gives our best estimate for the infinite stress density $n = (1.0 \pm 0.2) \times 10^{16}$ cm^{-3}. The uncertainty in this value arises from the poor fit to the data in Fig 8; the calculated curve for $n(S)$ can be seen to approach the high-stress limit more slowly than do the data. However, our extrapolation is supported by the agreement with the theory of Liu[25] at $S = 5.5$ kg/mm^2. From the value of n at $1/S = 0$, we can calculate E_{Fe} and E_{Fh} and we obtain $E_{Fe} = 0.8 \pm 0.1$ meV and $E_{Fh} = 1.9 \pm 0.2$ meV. All of these results are in excellent agreement with the theoretical values cited above for infinite stress, which have been reviewed by Rice.[26]

In the above analysis, illustrated in Fig. 6, the critical value of stress for decoupling the electron valleys explains the anomalies seen in luminescence as a function of stress in the early work which has been reviewed by Hensel, Phillips, and Thomas.[27] The smooth shape of the hole density of states, illustrated in Fig. 4, likewise explains the absence of similar anomalies when the lower hole band becomes depopulated.

In our analysis, we have not resolved the interesting question of the values of the e-h liquid ground-state energy E_G and the energy difference ϕ between the lowest exciton band edge and the e-h liquid chemical potential μ'. Our absolute photon energies for the liquid are not sufficiently accurate to allow a subtraction from independent measurements of the exciton energy positions. Feldman, Chou, and Wong[5] have made simultaneous measurements of exciton and liquid energies at $S = 13.5$ kg/mm^2 and find $\phi = 0.65 \pm 0.07$ meV. Based on changes that we expect (in agreement with the results of Ref. 6) in the energies at higher S due to the nonparabolic hole bands, we estimate a reduction of about 10% between $S = 13.5$ kg/mm^2 and the infinite-stress limit. Such a reduction suggests reasonable agreement with the theoretical predictions for high stress: exciton binding energy $E_X = 2.65$ meV,[21] $-E_G = 3.08$ meV,[22] and therefore $\phi = 0.43$ meV.

V. CONCLUSION

We have analyzed luminescence spectra from the e-h liquid in Ge at a series of compressive stresses along the [111] crystallographic axis. In obtaining energies and densities to compare with theoretical results in infinite stress, we have emphasized the fact that none of the existing measurements is very close to this limit. We have therefore extrapolated the results and find excellent agreement with the existing theoretical predictions for the electron and hole Fermi energies (factors

of 3.3 and 2.0 smaller than at zero stress) and for the density (a factor of 24 smaller).

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