Experimental Evidence for Phase Separation of a Two-Component Electron-Hole Liquid in ⟨111⟩-Stressed Ge

J. Bajaj, Fei-Ming Tong,[a] and George K. Wong
Department of Physics and Astronomy and Materials Research Center, Northwestern University, Evanston, Illinois 60201
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Measurements of time-resolved luminescence spectra of electron-hole liquid in Ge under ⟨111⟩ uniaxial stress are reported. It is shown that these spectra present strong evidence that a novel phase separation recently predicted theoretically for this system does indeed occur. The critical ratio x_c of the number of hot electrons to that of cold electrons below which the phase separation occurs is found to be 0.63 for a stress of 6.8 kg/mm^2.

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Ge has a fourfold-degenerate conduction band at L points of the Brillouin zone. Under ⟨111⟩ uniaxial stress, three of the conduction valleys shift to higher energy while the fourth shifts to lower energy. It has been shown experimentally[1,2] that the intervalley scattering time is rather long (~1 μs) so that in an electron-hole drop (EHD) in this system it is possible for a substantial fraction of the electrons to be present in the three upper valleys. Similar observation has also been reported for ⟨100⟩-stressed Si.[3] The intervalley scattering time in Ge is sufficiently long for the electrons in the upper valleys ("hot" electrons) to thermalize within the upper valleys and to be in quasiequilibrium with the electrons in the lower valley ("cold" electrons) and with the holes. Thus, EHD in ⟨111⟩-stressed Ge provides a *unique* opportunity to study experimentally the thermodynamics of a two-component degenerate Coulomb Fermi liquid. Recent theoretical calculations by Kirczenow and Singwi[4] predict that under appropriate ⟨111⟩ stress a novel phase separation can occur. *When the ratio x of the total number of hot electrons to that of cold electrons is larger than a critical value x_c, hot and cold electrons can coexist homogeneously in EHD; while for $x < x_c$, a separation into two coexisting phases with quite different x ratios is possible.* In this paper, we report measurements of time-resolved recombination luminescence spectra of EHD in ⟨111⟩-stressed Ge. A detailed analysis of the spectra corresponding to different ratios x reveals evidence that the predicted novel phase separation indeed occurs.

The basic idea of our experiment is as follows. With a laser pulse much shorter than the lifetime of hot electrons, it is possible to create EHD with a relatively large ratio x right after the excitation. With increasing time after the initial laser pulse, the ratio x decreases since the lifetime of hot electrons is shorter than that of cold electrons.[1,2] The luminescence spectra of EHD with different ratio x can thus be obtained by making measurements at various delay times. An important advantage of this method is that the same sample and stress conditions can be maintained throughout each run, avoiding uncertainties which can obscure the interpretation of our results. Also, because hot and cold electrons have quite different lifetimes, they could separate spatially during diffusion from the surface if Ge is surface excited. To avoid this, we used volume excitation in our experiment. This was achieved by using 1.9-μm laser pulses to optically excite Ge via two-photon absorption. The 1.9-μm laser pulses of 10 ns duration were obtained as stimulated Raman emission from H_2 gas pumped by a Q-switched Nd-YAlG (neodymium-doped yttrium aluminum garnet) laser operating at 1.06 μm and 10 pulses per second. The beam diameter was ~3 mm throughout the sample and the laser power was adjusted so that an effective absorption coefficient of about 1 cm^{-1} was achieved. Preparation of samples and the stress device has been described earlier.[1] A cooled Ge diode with a time constant of 150 ns was used to detect the EHD luminescence. The signal from the Ge diode was measured with a boxcar integrator with an aperture time of 100 ns. As a test of our experimental system, EHD luminescence spectra of Ge at zero stress and 2 °K were measured. The EHD luminescence had a lifetime of 42 μs and showed *identical* spectra for *all* delay times. Using the bath temperature of 2 °K, we obtained an excellent theoretical fit to this spectrum yielding an electron-hole pair density of 2.35×10^{17} cm^{-3}, which compares well with the values obtained from cw measurements.[5] The same laser-power level and laser-beam size were used in experiments under stress to assure that optical power

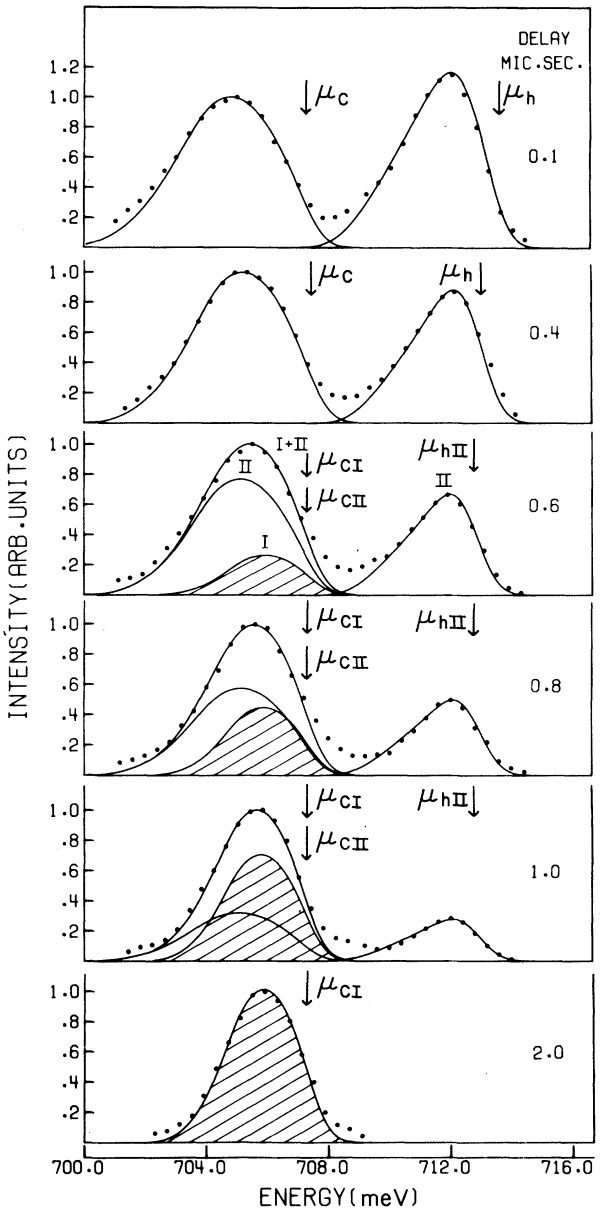

FIG. 1. Time-resolved luminescence spectra for Ge under a ⟨111⟩ uniaxial stress of 6.8 kg/mm^2 and at T = 2 °K. The points are data and the curves are calculated with use of the procedures described in the text. At each delay time, the peak intensity of the "cold" EHD line is arbitrarily normalized to 1. The μ's are the various spectroscopic chemical potentials determined from theoretical fits.

did not heat the sample above the bath temperature.

Figure 1 shows time-resolved luminescence spectra obtained at 2 °K and under a ⟨111⟩ stress of 6.8 kg/mm^2; similar results are obtained for other stresses favorable for phase separation and will be reported elsewhere. The line at lower (higher) energy is due to the recombination of cold (hot) electrons with the holes.[1] We shall refer to these lines as cold and hot EHD lines. It is observed that after a delay time of 0.4 μs the half-width of the cold EHD line narrows rapidly and the low-energy half-maximum position also moves rapidly to higher energy. Also, it should be noted that the peak position of the cold EHD line moves to higher energy as a function of time. This is important in that it rules out the possibility that the observed time evolution of the EHD spectrum is due to residual stress inhomogeneity since the stress gradient always makes the EHD line move to lower energy.[6]

We first analyze these spectra by assuming that hot and cold electrons coexist homogeneously within the same droplet. In this case, the hot and cold EHD line shapes are simply the convolution of hot- and cold-electron Fermi seas with the same hole Fermi sea. To a good approximation, we take the recombination matrix elements for hot and cold electrons to be the same. With this assumption, we have (with $N_e^h/N_e^c \equiv x$)

$$I_h/I_c = N_e^h/N_e^c = n_e^h/n_e^c, \tag{1}$$

where I_h and I_c are the experimentally determined integrated intensities of hot and cold EHD lines, N_e^h and N_e^c are the total numbers, and n_e^h and n_e^c are the total densities of hot and cold electrons, respectively. The condition for charge neutrality is

$$n_h = n_e^c + n_e^h, \tag{2}$$

where n_h is the hole density. Because of (1) and (2), the hole density n_h is the *single* adjustable parameter in this convolution procedure. Using the correct densities of states of ⟨111⟩-stressed Ge,[7] we obtained excellent theoretical fits for the spectra at delay times earlier than 0.4 μs, as can be seen from the top two curves in Fig. 1. However, this procedure failed at longer delay times. An example of the discrepancy between theoretical fit and experimental data is shown in Fig. 2(a). Even if one allows the recombination matrix element to be different for hot and cold electrons so that the theoretical cold EHD line can be scaled independently as shown by the dashed curve, the fit is still poor. We take this as an indication that phase separation has occurred and proceed to analyze the spectra accordingly.

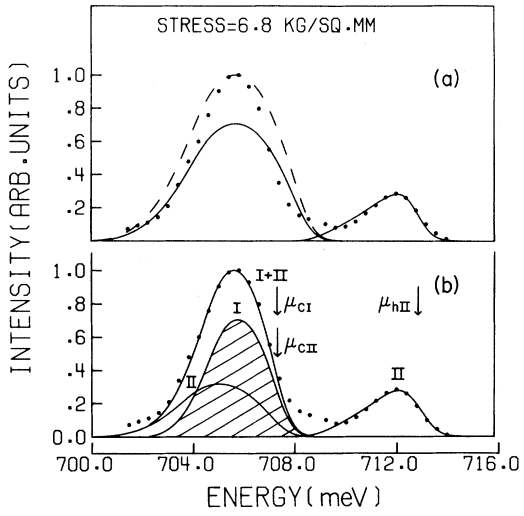

FIG. 2. Luminescence spectrum at 1.0 μs delay and at $T = 2$ °K. Points are data. (a) Solid curve is a theoretical fit obtained by assuming single phase. Dashed curve is explained in the text. (b) Solid curves are theoretical fits obtained by assuming phase separation. Curves I and II are line shapes due to phase I and II, respectively.

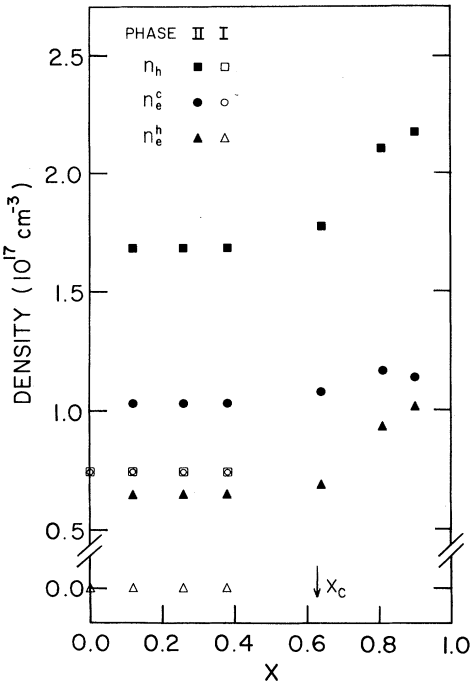

FIG. 3. Densities of hot electrons, cold electrons, and holes as a function of the average ratio x. x_c is the experimentally determined critical ratio.

After phase separation occurs, the experimental hot and cold EHD lines are the superposition of the contributions from both phases. Theoretical calculations show that at 2 °K one of these two phases (call it phase I) has a negligible amount of hot electrons so that essentially only the other phase contributes to the hot EHD line. This is also supported by the experimental observation that the line shape of the hot EHD line remains unchanged after a delay time of 0.4 μs. We shall assume that the phase I has no hot electrons. The other phase, which has the ratio $x = x_c$, will be called phase II. Experimentally, the cold EHD line shape due to phase I can be obtained by measuring the spectrum at long delay time when essentially all hot electrons have decayed and only phase I remains. The spectrum at 2.0 μs delay time (curve 6 in Fig. 1) should be an excellent approximation. Using the fact that, in phase I, the hole density is equal to the cold-electron density, we obtained an excellent theoretical fit to this spectrum, yielding an electron-hole-pair density of 7.3×10^{16} cm^{-3}. For delay times between 0.4 and 2.0 μs, phase II contributes significantly to the line shape. Its contribution can be obtained with the same procedure described earlier for the case of a single homogeneous phase with use of $x = x_c$. x_c should, of course, be the ratio x corresponding to the last spectrum that can be fitted with a single-phase model. Our results give $x_c = 0.63$. Using this value of x_c, we obtained the contribution of phase II to hot and cold EHD lines. Subtracting this theoretical line shape from the experimental one, we found that the resulting spectrum has the same shape and position as the spectrum due to phase I. Figure 2(b) shows the situation for 1-μs delay time. With the same $x_c = 0.63$, this procedure worked well for all the spectra measured at delay times longer than 0.4 μs. Curves 3 and 4 of Fig. 1 show the situations for 0.6- and 0.8-μs delay times. One sees that the phase separation model can explain the spectra at long delay times while a single-homogeneous-phase model cannot. We should point out three observations which provide additional consistency checks on our interpretation. First, from the fitting we determined that the cold electrons in phase I and phase II have the same chemical potentials as shown in Fig. 1. This should, of course, be the case if both phases are coexisting in equilibrium with each other. Second, as shown in Fig. 1, phase I grows relative to phase II. Third, the value of x_c which fits all spectra after 0.4 μs satisfactorily spans a narrow range between 0.6 and 0.64.

We summarize in Fig. 3 the densities of elec-

trons and holes as a function of the ratio x. As in theory,[4] x is defined as the average ratio. Before phase separation, x is determined in Eq.(1). After the phase separation, x should be given by

$$x = \frac{N_{e\mathrm{II}}{}^{h}}{N_{e\mathrm{I}}{}^{c} + N_{e\mathrm{II}}{}^{c}} = \frac{I_{h\mathrm{II}}}{I_{c\mathrm{II}} + I_{c\mathrm{I}} n_{h\mathrm{II}}/n_{h\mathrm{I}}}, \quad (3)$$

where $N_{e\mathrm{II}}{}^{h}$ is the total number of hot electrons in phase II and the other quantities are similarly defined. Using the results obtained from theoretical fittings, x can be easily evaluated with Eq. (3).

In conclusion, we have shown that time-resolved luminescence spectra obtained from ⟨111⟩-stressed Ge present evidence supporting the occurrence of a novel phase separation predicted by Kirczenow and Singwi.[4] We believe this to be the first experimental evidence of a phase separation of a mixture of two degenerate Fermi liquids[8] of an elementary kind. The experimentally measured value $x_c = 0.63$ at 6.8 kg/mm^2 is somewhat larger than the theoretical value $x_c = 0.5$ (Ref. 4) at the same stress. This discrepancy is probably due to the fact that the theoretical calculation[4] assumes that the exchange plus correlation energy does not depend on the details of the band structure and hence the ratio x. A recent model calculation by Combescot and Singwi[9] shows that the x dependence of the exchange and correlation energy also favors phase separation. This can affect the calculated value of x_c. Finally, we should mention that our experiment indicated that the phase separation can occur on the time scale of 100 ns. While the kinetics of the phase separation has yet to be investigated theoretically, a simple calculation based on the known electron-hole scattering times and Fermi temperature indicated that an electron's diffusion length is long compared to drop radius. This suggests that the occurrence of the phase separation on a time scale of 100 ns is definitely possible.

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[a]On leave from the Shanghai Institute of Technical Physics, Academia Sinica, Shanghai, People's Republic of China.

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