

and other experiments. In both of these cases, it is easily possible that a regular motion of vortex structures can be set up under appropriate flow conditions, which we speculate may be related to certain observations of regular and irregular orbital fluctuations.⁶

It is interesting to speculate on the outcome of a measurement of quantized vorticity in ³He-A by the Vinen⁸ vibrating-wire experiment or otherwise. At a Vinen wire the boundary condition will require \hat{l} to be radial and the phase may rotate by any integer number of units 2π ; a texture in, for instance, a cylinder can simply add or subtract 4π to this, so that any integer amount of vorticity is possible. However, the results might be chaotic in the absence of a field because of vortex textures throughout the liquid. With a field the wire can again have integer vorticity but in the surrounding liquid the 4π double vorticity is the most stable vortex line, consisting of a "core" which is a Fig. 1 texture of size $\sim R_s$, and a conventional outer region. Thus, one may tend to add or subtract *double* units.

In summary, the most important point to be made is that dissipation in this superfluid is qualitatively different from that in other superfluids and in most broken-symmetry systems, in that it can occur by the motion of textures (rather like "topological solitons") and not only by singularities of the order parameter. Thus, the property of superfluidity takes a very novel form in this case.

We wish to acknowledge discussions with W. F. Brinkman and M. C. Cross, and D. J. Thouless's suggestion that the Vinen experiment should be examined. We thank the Aspen Institute for hospitality during the preparation of this Letter.

¹P. W. Anderson, *Rev. Mod. Phys.* **38**, 298 (1966).

²P.-G. de Gennes, in *Collective Properties of Physical Systems*, edited by B. Lundqvist and S. Lundqvist (Academic, New York, 1973).

³G. Toulouse and M. Kléman, *J. Phys. (Paris), Lett.* **37**, 149 (1976). It was, of course, already proven that vorticity is not quantized in A [see, for example, R. Graham, *Phys. Rev. Lett.* **33**, 1431 (1974)], but the topological argument gives a new and very fundamental point of view.

⁴W. F. Brinkman and D. D. Osheroff, *Phys. Rev. Lett.* **32**, 584 (1974). See also P. W. Anderson and W. F. Brinkman, in *The Helium Liquids*, edited by J. G. M. Armitage and I. E. Farquhar (Academic, New York, 1975), p. 315.

⁵M. C. Cross and P. W. Anderson, in *Proceedings of the Fourteenth International Conference on Low Temperature Physics, Otaniemi, Finland, 1975* edited by M. Krusius and M. Vuorio (North-Holland, Amsterdam, Vol. 1, p. 29

⁶See J. C. Wheatley, *Rev. Mod. Phys.* **47**, 415 (1975), especially Sect. VII; also D. N. Paulson, M. Krusius, and J. C. Wheatley, *Phys. Rev. Lett.* **37**, 599 (1976).

⁷P. W. Anderson, in *The Many Body Problem*, edited by E. R. Caianello (Academic, New York, 1964).

⁸W. F. Vinen, *Proc. Roy Soc. London, Ser. A* **260**, 218 (1961).

Condensation of Optically Excited Carriers in CdS: Determination of an Electron-Hole-Liquid Phase Diagram

R. F. Leheny and Jagdeep Shah

Bell Telephone Laboratories, Holmdel, New Jersey 07733

(Received 29 November 1976)

We demonstrate formation of an electron-hole liquid in CdS by phase separation of optically excited carriers. Measurement of the electron-hole-liquid density as a function of pump intensity and sample temperature determines the liquid portion of the liquid-gas coexistence curve giving a low-temperature liquid density of $2 \times 10^{18} \text{ cm}^{-3}$ and $T_c = 55^\circ\text{K}$.

We present measurements which demonstrate the phase separation that occurs when an electron-hole liquid (EHL) forms by condensation from a less dense gas of excitation in CdS. These measurements allow construction of the liquid portion of the gas-liquid coexistence curve. Experimental identification of the EHL phase, bound by 13 meV, in highly excited CdS has been recently reported¹⁻³ and these measurements are in good

agreement with the calculations of Beni and Rice.⁴ The liquid chemical potential was found to be independent of pump intensity at 2°K ,¹ suggesting that phase separation was occurring but no other evidence for this phenomenon was provided. The new results reported here provide additional evidence for phase separation and establish that the critical temperature, above which the liquid does not condense, is $T_c \approx 55^\circ\text{K}$. In addition the low-

temperature liquid density is found to be $N_0 = 2 \times 10^{18} \text{ cm}^{-3}$, a value 20% smaller than that calculated by Beni and Rice.⁴ While no detailed calculations of the EHL properties at high temperature are available we find that the liquid density variation with sample temperature is in good agreement with a scaled version of the Ge phase diagram calculated by Reinecke and Ying.⁵ We take this result to indicate that the interactions important for condensation in CdS are not too different from the properties of EHL condensation in indirect-gap materials.

EHL formation creates an inversion of filled conduction-band states with respect to empty valence-band states resulting in large optical gain in a direct-gap semiconductor. The resultant amplification of luminescence can cause considerable distortion of the recombination spectra making luminescence studies unreliable for determining EHL properties.⁶ However, the *gain* spectra can be used directly to establish the distribution of excited states in the crystal. At 0°K the gain linewidth, measured from the high-energy edge to the low-energy edge of the gain spectrum, corresponds to the sum of the carrier Fermi energies, which is directly related to the carrier densities. In addition the energy difference between the high-energy edge and the free-exciton energy corresponds to the EHL binding energy. At higher temperatures, thermal excitation of the Fermi system modifies the distribution of carriers shifting the high-energy edge of the gain spectrum to lower energy, but the gain linewidth is still equal to the sum of the carrier Fermi energies. We have used these relationships to determine the EHL density in CdS by measuring gain linewidth as a function of pump intensity and sample temperature.

Gain spectra were obtained by the method of amplified spontaneous emission⁶ using N_2 laser excitation (100-kW peak power, 10^{-8} sec pulse-width, and 30 pulses per sec). Measurements of gain with pulsed lasers of low duty factor require $gl \geq 0.1$ where g is the net gain per unit length and l is the path length over which amplification occurs. The method of amplified spontaneous emission is particularly useful for low excitation intensities since small net gain can be compensated by longer excitation length. Data were obtained using a sensitive Vidicon system to record the entire spectrum simultaneously and the measurement corresponds to a time average over the pumping pulse.

Figure 1 illustrates luminescence spectra ob-

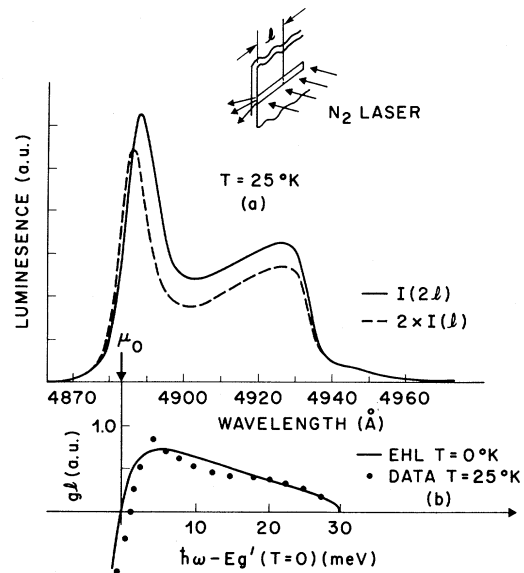


FIG. 1. (a) Spectra obtained for edge emission for two values of N_2 laser excitation length as shown in the inset. (b) Points correspond to logarithm of the ratio of spectra in (a) while solid line is a fit of the data assuming $N_0 = 2 \times 10^{18} \text{ cm}^{-3}$ and $T = 0^\circ\text{K}$. The sample temperature is 25°K , μ_0 corresponds to the chemical potential at $T = 2^\circ\text{K}$, and the small shift in chemical potential at 25°K is evident.

tained from the sample edge for two values of excitation length ($l \approx 6 \times 10^{-2} \text{ cm}$) and for a crystal temperature⁷ of 25°K with pump intensity of 10^4 W/cm^2 . These spectra contain exciton recombination features arising outside the liquid phase as well as contributions from the EHL. The two spectra intersect at an energy where loss changes to gain [$I(2l) = 2I(l)$] and the high-energy intersection corresponds to the chemical potential of the liquid phase, μ . The gain spectrum is determined from the logarithm of the ratio of these spectra $gl = \ln(R - 1)$. This gain spectrum results only from the condensed phase and has a typical EHL line shape as shown in the figure. This measurement of the liquid line shape agrees well with that determined from direct amplification of the probe beam at higher excitation intensity.¹ The solid curve is a fit of the gain line shape assuming parabolic bands and no momentum conservation in the electron-hole recombination.⁸

In Fig. 2(a) we summarize results of gain linewidth measurements made for a factor of 10^2 variation in pump intensity. The solid curve illustrates the expected linewidth for a plasma having an average density \bar{N} corresponding to the excitation density, taking the carrier lifetime to be 10^{-9} sec and excitation penetration depth of 10^{-4}

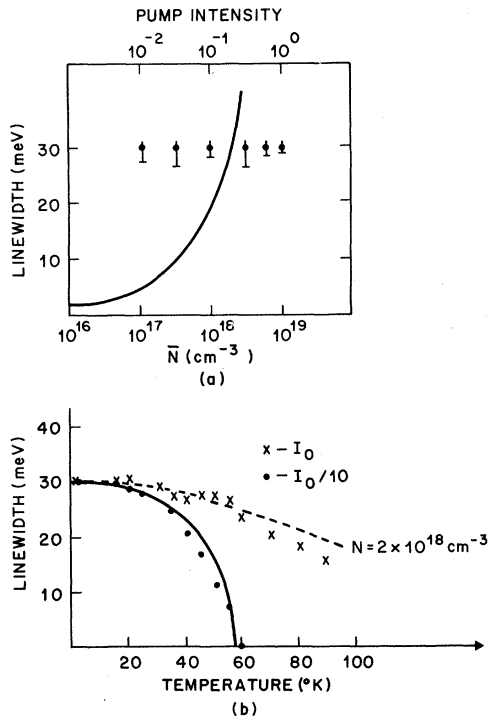


FIG. 2. (a) Variation in gain linewidth with pump intensity for sample temperature of 25°K. The solid points indicate the linewidth determined by fitting the data with a theoretical expression assuming no momentum conservation. The flag indicates the minimum linewidth estimated from the high- and low-energy cutoff of the measured gain. The solid curve indicates the variation of the sum of the carrier Fermi energies for the calculated average carrier density, \bar{N} . (b) Variation in gain linewidth with sample temperature for the values of pump intensity corresponding to \bar{N} above and below the low-temperature liquid density. Dashed curve, variation of constant density Fermi liquid; solid curve, phase diagram of Fig. 3.

cm. While these are only estimates of the actual lifetime and penetration depth, it is clear the measured linewidth is considerably broader at low pump intensity than expected for a plasma with density \bar{N} , and is insensitive to changes in pump intensity. The data are best described by a fixed density $N_0 = 2 \times 10^{18}$ cm⁻³. Condensation of the excitation is required to achieve this liquid density when $\bar{N} < N_0$ and for this condition the condensed phase is in equilibrium with the gas phase.

We can make use of this result to determine the temperature variation of the liquid phase that coexists with the gas phase by measuring the gain linewidth as a function of temperature. Typical results are shown in Fig. 2(b) for pumping conditions corresponding to $\bar{N} < N_0$ and $\bar{N} > N_0$. For $\bar{N} < N_0$ ($\bar{N} \approx 3 \times 10^{17}$ cm⁻³) the gain linewidth varies

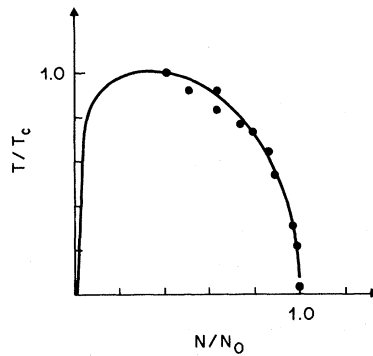


FIG. 3. Liquid-gas coexistence curve in the phase diagram of CdS. The solid line corresponds to the Reinecke-Ying calculation (Ref. 5) for Ge scaled for $N_0 = 2 \times 10^{18}$ cm⁻³ and $T_c = 55^\circ\text{K}$.

rapidly with temperature, with no measurable gain above $\approx 55^\circ\text{K}$. Since phase separation is occurring, a plot of liquid density variation represents the liquid portion of the gas-liquid co-existence curve and is shown in Fig. 3. Phase separation occurs up to some maximum temperature T_c above which only a single electron-hole-plasma phase with density \bar{N} exists. For the pump intensity used there is no gain resulting from this plasma for temperatures above T_c . We have estimated T_c for CdS by assuming that the exchange and correlation energies are independent of temperature and calculating the variation in chemical potential with density at each temperature.⁹ In this way we find $T_c \approx 50^\circ\text{K}$ in good agreement with our observation. At low temperatures our measured EHL density varies with increasing temperature as T^2 like a noninteracting Fermi liquid, while at higher temperature the decrease in liquid density is more rapid. Reinecke and Ying⁵ have calculated a phase diagram for Ge and Si and their result for Ge is in good agreement with the measurements of Thomas, Rice, and Hensel.¹⁰ In the absence of a detailed theory for the high-temperature behavior of an EHL in CdS we have compared our measurements to a scaled version of the Reinecke-Ying phase diagram for Ge shown as the solid line in Fig. 3.

For the case where $\bar{N} > N_0$ the pump laser can generate an excitation density greater than the liquid density and for this case we observe considerably less temperature variation of the gain linewidth. In this case the observed reduction of linewidth with increasing temperature can be accounted for by calculating the thermal variation of the sum of the electron and hole Fermi energies for constant carrier density. The dashed

curve in Fig. 2(b) illustrates the result of such a calculation for $N = 2 \times 10^{18} \text{ cm}^{-3}$ in CdS. Since the EHL has a rather small compressibility for compression to densities larger than N_0 the excitation is expected to expand into the crystal until this density is achieved as indicated by the data shown in Fig. 2(a). However, at higher temperatures expansion beyond N_0 to the equilibrium density at the sample temperature is not observed. There is evidence that in Si at higher temperatures expansion of an initially overdense plasma ($N \approx N_0$ but greater than the equilibrium density) takes a time on the order of $10\text{--}20 \times 10^{-9} \text{ sec}$.¹¹ This time is considerably longer than the excitation lifetime in a direct-gap material like CdS so it is unlikely that the equilibrium density can be achieved by expansion beyond N_0 during the excitation lifetime. These considerations suggest that the droplets of liquid expected to form for the case of underdense pumping can be expected to have rather small radius.

In conclusion we have investigated condensation of optically excited carriers in the direct-gap semiconductor CdS and find that the liquid phase can form from an underdense gas of excitation up to temperatures $\approx 55^\circ\text{K}$. These results determine the liquid portion of the gas-liquid coexistence curve for this system. While our understanding of condensation phenomena in the limit of pulsed, high-intensity excitation is not complete, we find that our results are in good agreement with similar measurements made under quite different pumping conditions for Ge, and indirect-gap material with very different excitation decay kinetics. The short lifetime of the excitation, and the high density of the liquid phase in CdS might be expected to create very different conditions governing the condensation process; nevertheless,

our results demonstrate that the factors determining the liquid portion of the coexistence curve for EHL's are quite general with similar behavior for materials having very different band structures.

¹R. F. Leheny and Jagdeep Shah, *Phys. Rev. Lett.* **37**, 871 (1976).

²V. G. Lysenko, V. I. Revenko, T. G. Tratas, and V. B. Timofeev, *Zh. Eksp. Teor. Fiz.* **68**, 335 (1975) [*Sov. Phys. JETP* **41**, 163 (1975)].

³G. O. Müller, in *Proceedings of the Taormina Research Conference on the Structure of Matter: Recent Developments in Optical Spectroscopy of Solids*, Taormina, Italy, 1976 (to be published), and private communication.

⁴G. Beni and T. M. Rice, *Phys. Rev. Lett.* **37**, 874 (1976).

⁵T. L. Reinecke and S. C. Ying, *Phys. Rev. Lett.* **35**, 311 (1975).

⁶K. L. Shaklee, R. E. Nahory, and R. F. Leheny, *J. Lumin.* **7**, 284 (1974).

⁷All temperatures reported are sample temperatures. For the pump intensity used carrier heating is not expected to be significant. [See J. Shah, *Phys. Rev. B* **9**, 562 (1974).]

⁸While this assumption gives a good fit to the data as in the case GaAs [E. Göbél, *Appl. Phys. Lett.* **24**, 492 (1975)] further investigation of the EHL line shape in direct-gap materials is required.

⁹The details of this calculation will be published in a longer article describing this work, but the calculation is essentially the same as the graphical construction used by M. Combescot [*Phys. Rev. Lett.* **32**, 15 (1974)] to determine T_c for Ge.

¹⁰G. A. Thomas, T. M. Rice, and J. C. Hensel, *Phys. Rev. Lett.* **33**, 219 (1974).

¹¹Jagdeep Shah and A. H. Dayem, *Phys. Rev. Lett.* **37**, 861 (1976).

Computer Model of Metallic Spin-Glasses

L. R. Walker and R. E. Walstedt
Bell Laboratories, Murray Hill, New Jersey 07974
 (Received 6 January 1977)

The properties of dilute Ruderman-Kittel-Kasuya-Yosida-coupled classical spins randomly embedded in an fcc lattice have been investigated by computer simulation. Results at $T = 0$ are given for spatial correlations, molecular-field distributions, susceptibility, and frequencies of elementary excitations. Treating the latter as bosons, we find quantitative agreement with measured specific-heat values. The susceptibility $\chi(0)$ is also in accord with experiment.

A characteristic property of experimental spin-glass systems is that the low-temperature specific heat is approximately linear in T and substan-

tially independent of concentration c over some considerable range of c .^{1,2} Attempts to explain this behavior on the basis of single spin excita-