VOLUME 43, NUMBER 14 PHYSICAL REVIEW LETTERS 1 OCTOBER 1979

Scaling Relations for Electron-Hole-Droplet Condensation in Semiconductors

T. L. Reinecke
Naval Research Laboratory, Washington, D. C. 20375

and

S. C. Ying
Department of Physics, Brown University, Providence, Rhode Island 02912

(Received 27 April 1979)

It is pointed out that scaling relations suggested previously between the critical and ground-state properties of electron-hole droplets lack theoretical justification; by detailed calculations for model systems with widely varying band structure, these relations are shown not to be satisfied. New scaling relations are proposed, and their existence is traced to systematic trends in the band structure and the exchange-correlation energy of the electron-hole system.

The condensation of electrons and holes from a low-density gaseous phase into a high-density plasma characterized by electron-hole droplets (EHD) has been studied extensively in Ge. The complete phase diagram in density n versus temperature T has been measured, and the critical values n_c and T_c determined to rather high accuracy.[1] Recently corresponding studies have been made for other semiconductors such as Si,[2] strained Ge,[3] and compound systems.[4,5] Experimental uncertainties are considerably larger for these latter systems.

Based on the principle of corresponding states, it has been suggested[6] that the liquid-gas–like condensation phase diagrams when scaled to n_c and T_c should have the same universal shape as that for the classical liquid-gas condensation; a corollary is that φ/k_BT_c (φ is the condensation energy), which determines the gas side of the phase boundary at low temperature, is a constant for various systems. In addition, the empirical relations $T_c/n_0^{1/2}=\text{const}$ and $n_c/n_0=\text{const}$, where n_0 is the ground-state density, have been suggested.[5,6] However, there seems to be little theoretical grounds for the existence of these scaling rules: The principle of corresponding states appropriate for classical gases is not expected to apply to EHD condensation because the de Boer parameter for the electrons and holes is typically of the order of 80, making the electron-hole system the most quantum fluid known.[6] In addition, within the effective-mass approximation, the Hamiltonians for different systems differ in band degeneracy, and their Coulomb interaction is different in form from the interaction for classical gases. Thus, one would expect systematic deviations from the principle of corresponding states and variations in the shape of the phase diagram depending on the band structure of the semiconductor under consideration.

We have made detailed calculations of the critical properties of EHD condensation in six model systems involving Ge and Si with varying uniaxial strain and verify these points explicitly. These calculations, made for model systems with widely varying band structure, constitute the most extensive study of the systematics of EHD condensation to date. From our study, we propose a new set of scaling relations, and in addition we trace their origin to systematic trends in the properties of the electron-hole system.

In Ge and Si, the strain first eliminates the conduction-band degeneracy and then splits the coupled hole bands. These systems are denoted (Table I) as $X[v_e; v_h]$, where $X=\text{Ge}$ or Si, and v_e and v_h are, respectively, the number of electron or hole bands occupied. The correlation energies of these systems have been calculated by Vashishta, Das, and Singwi[7] to high accuracy, including band anisotropy and multiple scattering between electrons and holes. Their results are used in the present calculation. Two theoretical approaches are used here to calculate the critical properties. One is the uniform-plasma approach,[8] and the other is the droplet fluctuation approach[9]; the latter includes critical fluctuations in an approximate way and gives the phase diagram for $T \gtrsim T_c/2$ as well as values for T_c and n_c. The critical values T_c and n_c calculated with both approaches are in good agreement with recent experimental results,[10] e.g., T_c from the droplet fluctuation approach agree with experimental value to within 5%. For our present purposes, we list in Table I the results for the ratios φ/k_BT_c, $T_c/n_0^{1/2}$, n_c/n_0, and also $\delta \equiv \sigma(0)/n_0^{2/3}k_BT_c$, where $\sigma(0)$ is the surface energy per unit area. This

TABLE I. Values of ratios of ground state and critical properties obtained by microscopic calculations using the droplet fluctuation approach and those in parentheses using the uniform-plasma approach. Parameters are given in text. Units for the third row are $10^{-9}\,{}^\circ\mathrm{K\,cm}^{+3/2}$ and for the sixth row are $10^{-2}\,{}^\circ\mathrm{K\,cm}^{+3/4}$.

	n_c/n_0	φ/k_BT_c	$T_c/n_0^{1/2}$	$\sigma(0)/n_0^{2/3}k_BT_c$	$\lvert\epsilon(n_0)\rvert/k_BT_c$	$\frac{\kappa T_c}{n_0^{1/4}}\left(\frac{\kappa}{\mu}\right)^{1/4}$	$\frac{(m_{de}m_{dh})^{1/2}}{m_{de}+m_{dh}}$	$10(C/A)^{1/2}$	$\frac{\sigma(0)}{n_0^{2/3}k_BT_c}\frac{1}{10(C/A)^{1/2}}$
Ge[4; 2]	0.290 (0.193)	3.07 (2.61)	14.2 (16.7)	0.529	10.3 (8.76)	2.03 (2.38)	0.488	1.35	0.392
Ge[1; 2]	0.284 (0.163)	1.04 (0.835)	18.3 (22.8)	0.387	10.9 (8.75)	1.97 (2.44)	0.485	1.07	0.362
Ge[1; 1]	0.288 (0.222)	1.75 (1.35)	27.6 (35.7)	0.337	12.3 (9.52)	1.87 (2.41)	0.453	0.815	0.413
Si[6; 2]	0.295 (0.150)	3.66 (2.96)	13.0 (16.1)	0.449	11.0 (8.88)	1.95 (2.42)	0.477	1.18	0.380
Si[2; 2]	0.321 (0.196)	2.67 (1.90)	16.0 (22.5)	0.362	11.8 (8.40)	1.94 (2.71)	0.499	1.01	0.358
Si[2; 1]	0.316 (0.173)	1.71 (1.37)	21.2 (26.3)	0.306	12.0 (9.68)	1.94 (2.40)	0.465	0.798	0.383

last parameter provides a measure[9] of the shape of the scaled liquid-gas-like phase diagram (n/n_c vs T/T_c) with larger values corresponding to a flatter top for the diagram. The results for these ratios from the two different theoretical approaches show the same trends between systems. The constancy of n_c/n_0 is reasonably well satisfied, but φ/k_BT_c, $T_c/n_0^{1/2}$, and δ show systematic variations as one goes from the unstrained to strained systems. Thus we conclude that the apparent success of the existing scaling relations is circumstantial.

From our study, we propose a new set of scaling relations. In addition to the existing relation $n_c/n_0=\mathrm{const}$, these relations are $\lvert\epsilon(n_0)\rvert/k_BT_c=\mathrm{const}$ and $(\kappa T_c/n_0^{1/4})(\kappa/\mu)^{1/4}=\mathrm{const}$, where $\epsilon(n_0)$ is the EHD ground-state energy per pair, κ the background dielectric constant, and μ the optically averaged electron and hole mass (in units of bare electron mass). From Table I the constancy of these two new ratios are seen to be satisfied to a remarkable degree.

We now demonstrate that the existence of these new relations can be traced to systematic trends in the exchange-correlation energy and the band structure of various EHD systems. The free-energy density can be written as $nf(n,T)=n[An^{2/3}+\epsilon^{xc}(n)-Dn^{-2/3}\tau^2]$, where $A=(3/10)(3\pi^2)^{2/3}\mu(m_{de}^{-1}+m_{dh}^{-1})(2R_x)$, $D=\frac{1}{2}(\pi/3)^{2/3}\mu^{-1}(m_{de}+m_{dh})(2R_x)$, and $\tau=k_BT/2R_x$; m_{di} are the total density-of-states effective masses, and the usual atomic units are used; $2R_x=e^2/a_x\kappa$, $a_x=\kappa\hbar^2/\mu e^2$. The first term is the zero-T kinetic energy, and $\epsilon^{xc}(n)$ is the exchange-correlation energy taken to be independent of T.[8] The T dependence is the leading term in k_BT/E_F from the kinetic energy.[8] For an inhomogeneous system (surface), the leading gradient expansion from the kinetic energy has the form $Cn^{-1}\lvert\nabla n\rvert^2$. Within the uniform-plasma approach, $\epsilon(n_0)$ and n_0 are determined by minimizing $f(n,0)$, and T_c and n_c are determined from the inflection point in the chemical potential $\mu=\partial[nf(n,T)]/\partial n$. The shape parameter is obtained from a simple variational calculation[10] with use of a parametric form for the surface density profile $n(\vec{r})$. It has been already noted[11,12] that ϵ^{xc} calculated for different semiconductors has an almost universal form when expressed in appropriate atomic units. In fact, we find for the model systems under consideration, $\epsilon^{xc}(n)$ and its derivatives for densities between n_0 and n_c are given very accurately by a one-term polynomial of the form $\epsilon^{xc}(n)=bn^p(R_x)$ all with the same b

TABLE II. Ratios formed from most recent experimental values. Results for Ge[4;2] are from Refs. 1 and 6; and those for Si[6;2] are from Ref. 2; those for strained Ge are from Ref. 3; and those for GaP and SiC are from Ref. 5. Units for the third column are 10^{-2} °K cm$^{3/4}$.

	$\lvert\epsilon(n_0)\rvert/k_BT_c$	n_c/n_0	$(\kappa k_BT_c/n_0^{1/4})(\kappa/\mu)^{1/4}$
Ge[4;2]	10.3 ± 0.7	0.31 ± 0.07	2.01 ± 0.2
Si[6;2]	11 ± 1.6	0.35 ± 0.03	1.97 ± 0.7
Strained Ge	9.2 ± 3.0	0.28 ± 0.1	1.80 ± 1.0
GaP	≈ 9.5		
SiC	≈ 11.0		≈ 2.0

and p.[13] For this choice of $\epsilon^{xc}(n)$, one obtains

$$|\epsilon(n_0)|/k_BT_c = f_1(p)[AD/(2R_x)^2]^{1/2}, \tag{1}$$

$$n_c/n_0 = f_2(p), \tag{2}$$

$$(\kappa T_c/n_0^{1/4})(\kappa/\mu)^{1/4} = bf_3(p)[AD/(2R_x)^2]^{-1/2}\ ^\circ\mathrm{K\ cm}^{3/4}, \tag{3}$$

$$\delta \equiv \sigma(0)/n_0^{2/3}k_BT_c = [|\epsilon(n_0)|/k_BT_c]f_4(p)(C/A)^{1/2}, \tag{4}$$

where $f_i(p)$ are smooth functions of p. It is now seen that the approximate scaling rules based on (1)–(3) exist because (i) $[AD/2R_x)^2]^{-1/2}$ is approximately constant and (ii) the single term $\epsilon^{xc}(n)$ is a good representation for a large group of semiconductors. The parameter $[AD/2R_x)^2]^{-1/2} \propto (m_{de}m_{dh})^{1/2}/(m_{de}+m_{dh})$ depends very weakly on the ratio m_{de}/m_{dh}, and, for example, for the six model systems considered here, it varies by $\leq 5\%$ from the average value of 0.478. To assess the effect of the single polynomial form of $\epsilon^{xc}(n)$, we have computed all of the scaling ratios using the same uniform-plasma approach for the choice $b = 3.5$ and $p = 0.24$. This gives $|\epsilon(n_0)|/k_BT_c = 8.38$, $n_c/n_0 = 0.203$ for Ge[4;2] and $|\epsilon(n_0)|/k_BT_c = 9.02$ and $n_c/n_0 = 0.203$ for Ge[1;1]. The corresponding values in Table I calculated with the detailed correlation energy from Ref. 7 are 8.76, 0.193 for Ge[4;2] and 9.52, 0.222 for Ge[1;1]. Results for other systems are similar. Together with the weak variation in the parameter $[AD/2R_x)^2]^{-1/2}$, this indicates that the scaling relations (1)–(3) should hold for a wide variety of EHD systems with deviations between systems of the order of 20%. The parameter C/A, on the other hand, varies systematically with band structure. (For isotropic bands with degeneracy v as $v^{2/2}$.) This leads to variations in the shape parameter δ and a deviation from the universal shape of phase diagram[14] by as much as 40% for $T/T_c \sim 0.9$ and is consistent with the results in Table I.

Very recently EHD have been observed in a number of compound semiconductors.[4,5] In these systems, the exchange-correlation energy to which the electron-phonon interaction makes some contribution is not as well known; nonetheless, recent calculations[15] for GaP, GaAs, CdS, AgBr, and several other compound systems can be fitted between n_0 and n_c with the single polynomial form (with the same b and p as above) to within a few percent ($\leq 5\%$), and thus the relations (1) and (2) are expected to hold. These calculations[5,15] suggest that for many compound systems, a good approximation for calculating ϵ^{xc} is to scale the Coulomb interaction by the low-frequency dielectric constant ϵ_0 and use the polaron masses (systems with $E_F, \hbar\omega_p \leq \hbar\omega_{LO}$, where ω_p and ω_{LO} are the plasmon and LO-phonon frequencies); if this is the case then the relation based on Eq. (3) holds with ϵ_0 and polaron masses.

The values of the present scaling ratios are in agreement with available experimental results as summarized in Table II. At present, there is too little data to provide a definitive experimental test. More accurate measurements, especially for more widely varying systems would clearly be desirable.

One of us (T.L.R.) wishes to thank the Max-Planck-Institut, Stuttgart, for hospitality during the completion of this work, and the other (S.C.Y.) acknowledges support in part by National Science Foundation Grant No. DMR76-10376 and by the Material Research Laboratory Program at Brown University.

---

[1] G. A. Thomas, J. B. Mock, and M. Capizzi, Phys. Rev. B 18, 4250 (1978); W. Miniscalco, C.-C. Huang, and M. B. Salamon, Phys. Rev. Lett. 39, 1356 (1977).

[2] J. Shah, M. Combescot and A. H. Dayem, Phys. Rev. Lett. 38, 1497 (1977); A. Forchel and W. Schmid, unpublished.

[3] B. J. Feldman, H. L. Chou, and G. K. Wong, Solid State Commun. 24, 521 (1977).

[4] J. Shah, R. F. Leheny, W. R. Harding, and W. R. Wright, Phys. Rev. Lett. 38, 1164 (1977).

[5] D. Bimberg, M. S. Skolnick, and L. M. Sander, Phys. Rev. B 14, 2231 (1979); D. Bimberg, M. S. Skolnick, and W. J. Choyke, Phys. Rev. Lett. 40, 56 (1978).

[6] J. C. Hensel, T. G. Phillips, and G. A. Thomas, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1977), Vol. 32; G. A. Thomas, T. M. Rice, and J. C. Hensel, Phys. Rev. Lett. 33, 219 (1974).

[7] P. Vashishta, S. G. Das, and K. S. Singwi, Phys. Rev. Lett. 33, 911 (1974).

[8] M. Combescot, Phys. Rev. Lett. 32, 15 (1974); T. M. Rice, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1977), Vol. 32.

[9] T. L. Reinecke and S. C. Ying, Phys. Rev. Lett. 35, 311, 547(E) (1975), and Phys. Rev. B 13, 1850 (1976); a numerical error has been corrected.

[10] T. L. Reinecke, M. C. Lega, and S. C. Ying, Phys. Rev. B (to be published).

[11] See, e.g., G. Kirczenow and K. S. Singwi, Phys. Rev. Lett. 41, 326 (1978).

[12] The reason for this is not yet known in detail. Crudely $\epsilon^{xc}(n)$ is given to good approximation in terms of a single plasmonlike excitation and then scales roughly like the atomic unit of energy.

[13] For all six model systems the detailed values of $\epsilon^{xc}(n)$, $n\epsilon^{xc\prime}(n)$, $n^2\epsilon^{xc\prime\prime}(n)$, and $n^3\epsilon^{xc\prime\prime\prime}(n)$ for all densities between n_0 and n_c vary from those given by the single-polynomial form by at most 25%, 5%, 14%, and 17%, respectively.

[14] Recent evidence (Thomas, Mock, and Capizzi, Ref. 1) suggests that for Ge[4; 2] dissociation in the gas may affect the shape of the gas side of the diagram, but the situation is not yet clear. If this occurs then "shape" used here refers to the curvature of the liquid side.

[15] G. Beni and T. M. Rice, Phys. Rev. B 18, 768 (1978); L. V. Keldysh and A. P. Silin, Zh. Eksp. Teor. Fiz. 69, 1053 (1975) [Sov. Phys. JETP 42, 535 (1976)]; G. Beni, T. M. Rice, and L. M. Hemstreet, to be published.

ERRATA

JETS PRODUCED IN π^-, π^+, AND PROTON INTERACTIONS AT 200 GeV ON HYDROGEN AND ALUMINUM TARGETS. C. Bromberg, G. Fox, R. Gomez, J. Pine, J. Rohlf, S. Stampke, K. Yung, S. Erhan, E. Lorenz, M. Medinnis, P. Schlein, V. Ashford, H. Haggerty, R. Juhala, E. Malamud, S. Mori, R. Abrams, R. Delzenero, H. Goldberg, S. Margulies, D. McLeod, J. Solomon, R. Stanek, A. Dzierba, and W. Kropac [Phys. Rev. Lett. 42, 1202 (1979)].

Reference 8 should read K. Yung, thesis, California Institute of Technology, 1979 (unpublished).

LEPTON MASS FORMULA. A. O. Barut [Phys. Rev. Lett. 42, 1251 (1979)].

Reference 1 should be expanded to read as follows:

[1] A. O. Barut, Phys. Lett. 73B, 310 (1978), and references cited therein [in particular, Y. Nambu, Progr. Theor. Phys. 7, 595 (1952); G. Rosen, Nuovo Cimento 32, 1037 (1964)].