PHYSICAL REVIEW B VOLUME 26, NUMBER 12 15 DECEMBER 1982

Exciton-plasma Mott transition in Si

George B. Norris and K. K. Bajaj*
Air Force Wright Aeronautical Laboratories, AFWAL/AADR, Wright-Patterson Air Force Base, Ohio 45433
(Received 10 June 1982)

The transition between free excitons and an electron-hole plasma is described for the first time by a theory based on full random-phase-approximation (RPA) screening in the plasma. The onset of exciton binding predicted is in excellent agreement with experimental data attributed to exciton dissociation in Si. It is suggested that the many-body electron-hole interaction may be well described by static RPA screening near the Mott transition.

I. INTRODUCTION

The behavior of optically pumped electron-hole pairs in indirect-gap semiconductors has been extensively studied in the last decade. The formation of electron-hole drops (EHD), which arises from a first-order liquid-gas transition between the electron-hole fluid and exciton gas, is well established and understood.[1] However, the Mott transition which must take place between the insulating free exciton (FE) gas and the metallic electron-hole plasma (EHP), particularly above the critical temperature T_c for the liquid-gas transition, is presently far from being well understood. While such a transition would be quite diffuse for thermal and entropy ionization processes, a much sharper transition arising from many-body effects (screening) is thought to occur at lower temperatures and higher densities.[2,3] In this paper we examine experimental data[4,5] associated with exciton dissociation in silicon and show that the most general form of random-phase-approximation (RPA) screening for arbitrarily degenerate plasmas yields excellent agreement with the experimental results of Refs. 4 and 5.

II. EXPERIMENT

In photoluminescence studies of Si (Refs. 4–7) and Ge (Ref. 8), the free-exciton line is seen to broaden on the low-energy side when pumped beyond a certain intensity. Above T_c, the broadening evolves into a shifting peak, which, for sufficient pumping intensity, is well fitted by an electron-hole plasma line shape. Below T_c, the broadening occurs quite near the liquid-gas transition and the resulting spectra are complicated by luminescence from electron-hole drops. Thomas and Rice[9] interpret the broadening below T_c as being due to the formation of trions (charged excitons) and biexcitons. Forchel *et al.*[6] explain the broadening in terms of electron-hole plasma luminescence, based on the observed behavior under stress. Above T_c, the continuously shifting behavior would seem to be consistent with the formation of an electron-hole plasma. In this connection, it has been noted that the onset of the line broadening agrees roughly with the Mott criterion

$$n_{\text{Mott}}=(q_{\text{DH}}^2\epsilon_0 k_B/8\pi e^2)T\ , \qquad (1)$$

further suggesting that the broadening and shift are associated with a Mott transition between free excitons and an electron-hole plasma. Here n_{Mott} is the density of electron-hole pairs at the transition, ϵ_0 is the static dielectric constant, and q_{DH} is the Debye-Hückel screening wave vector for a classical plasma, evaluated where the binding energy of a free exciton with a statically screened electron-hole potential goes to zero. Numerical evaluation[10] of q_{DH} for parameters appropriate to silicon ($\epsilon_0=11.4$ and exciton reduced mass $m_{\text{ex}}=0.123m_e$) gives

$$n_{\text{Mott}}=(1.60\times10^{15}\ \text{cm}^{-3}/\text{K})T\ , \qquad (2)$$

which is only a factor of 2 lower in density than the relevant experimental data[4,5] (see Fig. 1). However, it must be pointed out that absolute experimental determination of the gas density is not currently possible.[5,6] Shah *et al.*[4] assume a temperature-independent linear scaling between pumping intensity and electron-hole-pair density. This relation is scaled by relating the appearance of EHD luminescence to the gas density on the liquid-gas coexistence curve of Reinecke and Ying,[11] which is fitted to an experimentally determined critical temperature of $T_c=27$ K and $T=0$ liquid density of $n_0=3.3\times10^{18}$ cm^{-3}. Recently, Forchel *et al.*[5] have performed a similar analysis of independently measured luminescence data. Their analysis differs by the use of $T_c=23$ K and $n_0=3.15\times10^{18}$ cm^{-3}

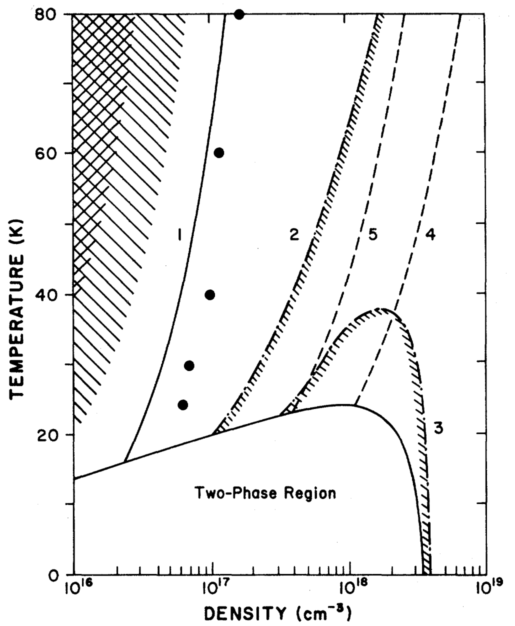

FIG. 1. Density-temperature phase diagram of electron-hole pairs in unstressed silicon. The area labeled "two-phase region" is rendered unobservable by the phase separation between electron-hole drops and low-density gas (exciton or plasma). The Debye-Hückel Mott criterion is shown as curve 1 and may be compared to the experimental data in Ref. 5, shown as solid circles. The singly shaded region indicates where the electron plasma behaves classically and the doubly shaded region is for classical behavior of holes as well. Dynamical screening becomes important for electrons to the right of curve 2 and for holes below curve 3. Curves 4 (for electrons) and 5 (for holes) indicate the intermediate region between degenerate and nondegenerate plasmas as given by the equality between the chemical potential and k_BT.

(in agreement with theory and experiment) and by a more sensitive measure of line broadening. The effects of these refinements on the experimental gas density tend to offset each other, and give data very similar to that of Shah *et al.* While such calibration procedures are of somewhat uncertain accuracy, their use is currently unavoidable and represent the best available estimate of the Mott-transition densities.

III. THEORY

Theoretical work on the Mott transition has generally followed two approaches: evaluation of a simplified thermodynamical model of the FE-EHP-EHD system[2,3,12,13] or detailed calculation of the many-body exciton binding energy.[14–16] Neither approach has been entirely satisfactory to explain the available data due to inherent approximations or simplifying assumptions made in the course of calculation. Our approach will be to generalize the Mott criterion described above taking into account the band structure and degree of degeneracy in the plasma, while remaining within the approximation of using a statically screened electron-hole potential. In this we are motivated by the observation that Debye-Hückel screening, strictly valid only for completely nondegenerate (classical) plasmas, overestimates the electron-hole binding-energy reduction afforded by screening in an arbitrarily degenerate plasma. (Note that in Fig. 1 the data fall well outside the region where purely classical screening can be assumed to apply.) Therefore, a general treatment of screening should tend to move the onset of exciton binding to higher densities, in agreement with experimental observations (see Fig. 1). As we consider screening by a plasma, rather than free excitons, we cannot calculate the screened exciton binding energy, but rather determine the onset of exciton binding as approached from the plasma side of the Mott transition.

In silicon, the center of mass exciton Hamiltonian can be written as[17]

$$\hat{H}_{\mathrm{ex}} = (\hbar^2/m_{\mathrm{ex}})\nabla^2 + V(r) + \hat{H}_d \;, \tag{3}$$

where m_{ex} is the optical reduced mass

$$m_{\mathrm{ex}}^{-1} = \tfrac{1}{3}(2/m_{\mathrm{et}} + 1/m_{\mathrm{el}}) + \gamma_1 \;, \tag{4}$$

with transverse and longitudinal electron masses m_{et} and m_{el} and the valence-band Kohn-Luttinger parameter γ_1. Here r is the electron-hole separation, $V(r)$ is the screened electron-hole potential, and the H_d contains the anisotropic and degenerate portions of the band structure, generally taken as a perturbation. The screened potential is most simply expressed in terms of its Fourier transform

$$V(q) = -4\pi e^2/q^2\epsilon(q) \;, \tag{5}$$

where $\epsilon(q)$ is the dielectric function for the plasma. In what follows, we will neglect H_d, as this has been shown to have a relatively small effect on the exciton binding energies,[17] and the conduction-band anisotropy is known to give results similar to the isotropic case in simple donor screening.[18] The dielectric function is taken in the static random-phase approximation for an isotropic electron-hole plasma of density n and temperature T:

$$\epsilon(q) = \epsilon_0 + \frac{4\pi n e^2}{k_B T_q} \sum_i \frac{G(x_i,\eta_i) F_{-1/2}(\eta_i)}{F_{1/2}(\eta_i)} \;, \tag{6}$$

where

$$x_i = \hbar^2 q^2 / 2m_i^* k_B T \;, \tag{7}$$

$$\eta_i = \mu_i / k_B T \ , \tag{8}$$

and

$$G(x,\eta) = [(\pi x)^{1/2} F_{-1/2}(\eta)]^{-1} \times \int_0^\infty dz\,(e^{z-\eta}+1)^{-1} \times \ln \left| \frac{1+\frac{1}{2}(x/z)^{1/2}}{1-\frac{1}{2}(x/z)^{1/2}} \right| . \tag{9}$$

In the summation, $i=1$ is for electrons and $i=2$ is for holes. Here F_j is the Fermi integral of order j, m_i^* is the electron (hole) optical mass, m_i is the electron (hole) density of states mass, ν_i is the conduction- (valence-) band degeneracy, and μ_i is the electron (hole) chemical potential given by

$$F_{1/2}(\eta_i) = \frac{n}{2\nu_i} \left[\frac{2\pi}{m_i k_B T} \right]^{3/2} . \tag{10}$$

For ease of computation, we use a polynomial interpolation for $G(x,\eta)$ developed by Meyer[19] that is accurate to 1%. We evaluate the binding energy by the use of a Hulthén 1s wave function as a variational trial state,[20]

$$\psi = \tfrac{1}{2}(\pi a)^{-1/2} \left[\frac{4}{\mu^2} - 1 \right]^{1/2} r^{-1} \times (e^{-[1-(1/2)\mu]r/a} - e^{-[1+(1/2)\mu]r/a}) \ ,$$

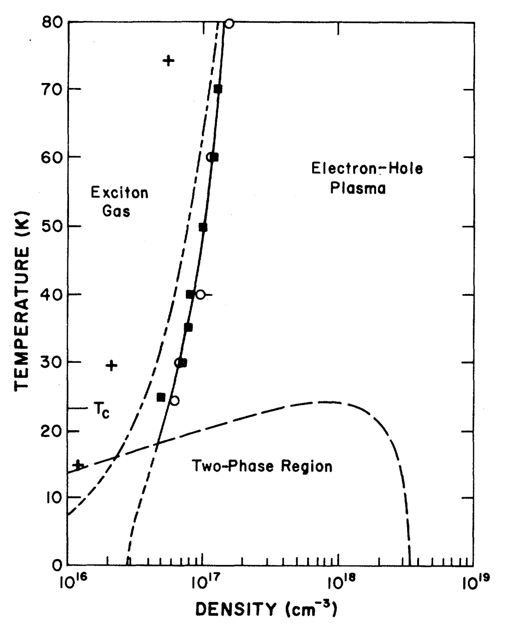

FIG. 2. Mott transition in unstressed silicon. The Debye-Hückel Mott criterion is given by the doubly dashed line. The results of Ref. 15 are shown as crosses. The results of the present work are shown as the solid line. These may be compared to the experimental results of Ref. 4 (solid squares) and Ref. 5 (open circles).

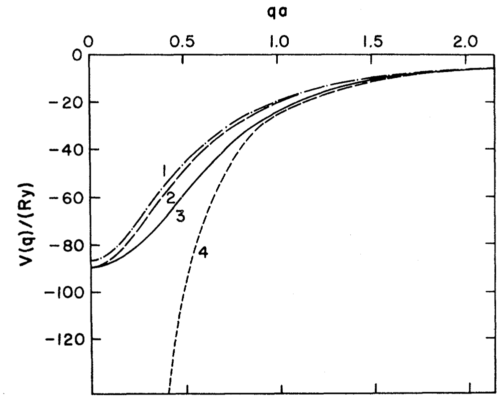

FIG. 3. Effective electron-hole interaction near the Mott transition ($T=31.2$ K, $n=7\times10^{16}$ cm^{-3}). Curve 1 is the Debye-Hückel result. Curve 2 is the result for the small wave-vector approximation to the RPA result. Curve 3 is the full RPA potential. Curve 4 is the unscreened potential.

where $a=\epsilon_0\hbar^2/e^2m_{\rm ex}$ is the exciton Bohr radius and μ is a variational parameter. The Hulthén wave function is an eigenstate of a short-range potential and has been shown to be a considerable improvement over hydrogenic trial states, with errors of less than 1–2% when compared to exact numerical solutions of several screened potentials.[21]

IV. RESULTS AND DISCUSSION

Using parameters for unstressed silicon ($m_{\rm et}=0.1905m_e$, $m_{\rm el}=0.916m_e$, $\gamma_1=4.28/m_e$, $\nu_e=6$, $\nu_h=2$, and $\epsilon_0=11.4$), we have solved for the density and temperature where the onset of exciton binding occurs. As shown in Fig. 2, this results in excellent agreement with the best available experimental data. The improvement over the Debye-Hückel result is entirely due to the use of the most general form of the RPA dielectric function. As shown in Fig. 3, for a density and temperature near the Mott transition the potential for RPA screening (curve 3) is considerably wider and somewhat deeper than the Debye-Hückel potential (curve 1), giving rise to the increased binding observed. The Dingle-Mansfield potential [small q limit of the RPA given by $G(x,\eta)\to1$ and shown in curve 2] is not appreciably different from the Debye-Hückel case and is insufficient to reproduce the results found. The results found are fitted numerically by

$$n_{\rm Mott} = (0.1260e^{-0.1302T} + 0.2414 + 0.01641T) \times 10^{17}$$

in units of cm^{-3} (T in K), to within the estimated accuracy of the calculation (5%). We have also determined the Mott transition for parameters appropriate to uniaxially stressed silicon.[22] Denoting the band structure with ν_e occupied conduction minima and ν_h occupied valence maxima by $[\nu_e;\nu_h]$, the [6;1], [4;1], and [2;1] stressed band configurations yield very similar results (less than 10% variation in density) to the unstressed [6;2] configuration. This small dependence on band occupancy can be traced to the relatively small number of carriers, which makes the plasma only weakly degenerate at higher temperatures and tends to make all carriers effective in screening [via the cutoff in $G(x,\eta)$ rather than the usual screening wave-vector cutoff] at lower temperatures.

These results follow from the approximation of replacing the many-body electron-hole interaction with a static Hartree effective interaction. This is a simplification in three major respects. Firstly, the dynamical nature of the interaction arising from inertial effects in the plasma have been neglected by assuming a statically screened effective two-particle interaction. This requires that the screening involve momentum transfers insufficient to excite real plasmons and introduce retarded interactions between electrons and holes (by coupling the single-particle and collective modes of the plasma). We quantify this by requiring that the screening wave-vector cutoff falls well below that required to excite real plasmons[23]

$$\omega_p \gg \hbar q_s^2/2m_i \ ,$$

which for the RPA is

$$1 \gg \frac{e^2 m_{\rm ex} m_i \nu_i^2 k_B T}{2\pi\epsilon_0 \hbar^4 n} F_{-1/2}(\eta_i) \ . \qquad (12)$$

As shown in Fig. 1, this condition is well satisfied for both electrons (curve 2) and holes (curve 3) at the densities and temperatures at the Mott transition. Secondly, the Hartree potential completely ignores exchange except for statistical occupation. This could be expected to be particularly important for the exciton as the screening carriers are identical with the particles being screened. However, as shown in Fig. 1, the Mott transition takes place in a very weakly degenerate region where thermal energies significantly exceed the electron and hole Fermi energies (note where the chemical potentials equal $k_B T$ as shown in curves 4 and 5). Thus, higher-order exchange effects could be thermally obscured. Thirdly, the Hartree potential ignores detailed correlations in the plasma. However, as for exchange, thermal effects could obscure the detailed quantum correlations in the plasma.

We conclude that excellent agreement can be achieved between the Mott criterion with RPA screening and experimental data attributed to the Mott transition in silicon. We emphasize that the theory involves no adjustable parameters. While the experimental data does reflect some uncertainties due to determination of electron-hole density, it is suggested that the effective electron-hole potential could be well described by RPA screening at the densities and temperatures near the Mott transition.

ACKNOWLEDGMENTS

We thank the authors of Ref. 5 for providing us with their experimental results in advance of publication and Dr. T. L. Reinecke for discussions of that work. We also thank Dr. J. Shah for discussions of experimental data and Dr. J. R. Meyer for providing us with his numerical interpolation of the RPA dielectric function. This work was supported in part by the Air Force Office of Scientific Research (AFOSR) under Contract No. F33615-81-C-1406.

---

*Present address: University Research Center, Wright State University, Dayton, Ohio 45435.

[1]H. 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, p. 87; T. M. Rice, *ibid.* p. 1.

[2]W. Ebeling, W. D. Kraeft, D. Kremp, and K. Kilimann, Phys. Status Solidi B 78, 241 (1976); W. D. Kraeft, K. Kilimann, and D. Kremp, *ibid.* 72, 461 (1975).

[3]H. Schweitzer, A. Forchel, and W. Klingenstein, Phys. Status Solidi B 102, 343 (1980).

[4]Jagdeep Shah, M. Combescot, and A. H. Dayem, Phys. Rev. Lett. 38, 1497 (1977).

[5]A. Forchel, B. Laurich, A. Hangleiter, W. Schmid, and T. L. Reinecke (unpublished).

[6]A. Forchel, B. Laurich, J. Wagner, W. Schmid, and T. L. Reinecke, Phys. Rev. B 25, 2730 (1982).

[7]A. F. Dite, V. G. Lysenko, and V. B. Timofeev, Phys. Status Solidi B 66, 53 (1974).

[8]G. A. Thomas, J. B. Mock, and M. Capizzi, Phys. Rev. B 18, 4250 (1978).

[9]G. A. Thomas and T. M. Rice, Solid State Commun. 23, 359 (1977).

[10]F. J. Rodgers, H. C. Graboski, and D. J. Harwood, Phys. Rev. A 1, 1577 (1970).

[11]T. L. Reinecke and S. C. Ying, Phys. Rev. Lett. 35,

311 (1975); 35, 547(E) (1975). A more recent reference is T. L. Reinecke, M. C. Lega, and S. C. Ying, Phys. Rev. B 20, 1562 (1979).

[12]G. Mahler and Joseph L. Birman, Phys. Rev. B 16, 1552 (1977).

[13]L. M. Sander and D. J. Fairobent, Solid State Commun. 20, 631 (1976).

[14]J. G. Gay, Phys. Rev. B 4, 2567 (1971); 6, 4884 (1972).

[15]R. Zimmerman, K. Kilimann, W. D. Kraeft, D. Kremp, and G. Röpke, Phys. Status Solidi B 90, 175 (1978).

[16]H. Haug and D. B. Tran Thoai, Phys. Status Solidi B 85, 561 (1978).

[17]N. O. Lipari and A. Baldereschi, Phys. Rev. B 3, 2497 (1971).

[18]C. Aldrich, Phys. Rev. B 16, 2723 (1977).

[19]J. R. Meyer, Ph.D. thesis, Brown University, 1977 (unpublished).

[20]L. Hulthén, Ark. Mat. Astron. Fiz. A 28, (5), 1 (1942).

[21]R. L. Greene, C. Aldrich, and K. K. Bajaj, Phys. Rev. B 15, 2216 (1977).

[22]J. C. Hensel and G. Feher, Phys. Rev. 129, 1041 (1963).

[23]This condition is essentially that given by Mahler and Birman (Ref. 12, p. 1558) as $k_c^2 R_{e,h}^2 \rightarrow 0$.