Investigation of Exciton-Plasma Mott Transition in Si

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Measurements of luminescence spectra in pure Si demonstrate the dissociation of excitons with increasing excitation intensity. The density at which the dissociation begins and its temperature variation agree well with Mott's predictions. We have also determined a phase diagram of Si, including the effects of electron-hole–liquid and Mott transition.

The Mott transition and other metal-insulator transitions have received considerable attention in recent years.[1] Although such a transition has been widely discussed for the exciton-plasma system in semiconductors, no experimental evidence has been reported. We report measurements of luminescence spectra in pure Si as a function of excitation intensity and temperature, which provide the first observation of the onset of dissociation of excitons into plasma. The magnitude and the temperature dependence of the exciton density n_D at which dissociation begins agree well with Mott's predictions. From our results we have also determined for the first time a phase diagram of Si, including the effects of electron-hole–liquid (EHL) and Mott transition. Within our experimental accuracy, we find no evidence for a second critical point[2] due to Mott transition. The possible effects of Mott transition on the phase diagram in Ge has been discussed by Thomas and co-workers.[3] Timusk[4] has also discussed effects of Mott transition in Ge, but has presented data only on thermal dissociation of excitons.

A crystal of pure Si, in the form of a Weierstrass sphere (radius ≈ 7mm), was excited by a 15-nsec pulse from a cavity-dumped Ar$^+$ laser (5145 Å) every 10 μsec. The temperature of the crystal, mounted in a Varitemp Dewar, was monitored by a Ge thermometer soldered to it and the luminescence signal was measured with gated (10 nsec) electronics.[5] We present below the results obtained at various excitation intensities I for a fixed delay (70 nsec from the beginning of the laser pulse); similar results are obtained as a function of delay at fixed I and will be reported elsewhere.

Figure 1 shows typical luminescence spectra[6] at various intensities I and temperatures T. At low I (curves 1) only one line is observed, characteristic of free-exciton (FE) luminescence, and the line shape is independent of I. For intermediate values of I (curves 2) the spectra depend on T: At 18 K a broad feature characteristic of EHL appears at lower energy while at 23 and 30 K the low-energy edge of the exciton line becomes broader. At still higher I (curves 3), one finds that at 23 K a second peak due to EHL appears at lower energy, while at 30 K a broad, almost symmetrical line, characteristic of an electron-hole plasma, is obtained. This behavior is best illustrated by plotting in Fig. 2 the position of the low-energy half-maximum[7] of every distinct peak in

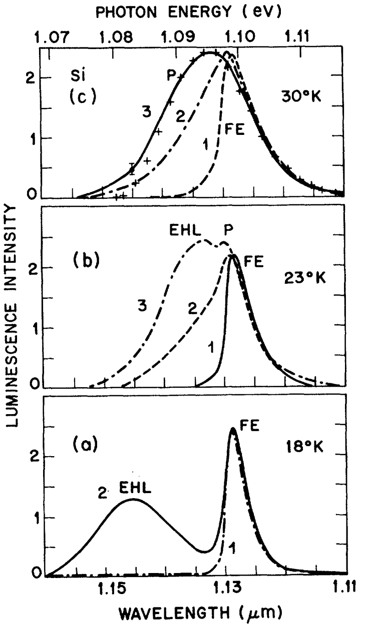

FIG. 1. Luminescence spectra for different P_a and T. P_a is the average incident power focused to ~ 100-μm spot. The values of P_a used are (a) 0.05 and 1 mW for curves 1 and 2, respectively; (b) 0.05, 0.4, and 2 mW, and (c) 0.05, 1, and 4 mW, for curves 1, 2, and 3, respectively. The resolution is 1 meV. Free-exciton (FE), plasma (P), and electron-hole-liquid (EHL) peaks are indicated. Experimental error bar is indicated in (c). The crosses represent fit to a calculated plasma line shape with $n = 3 \times 10^{18}$ cm^{-3} and $T = 30$ K.

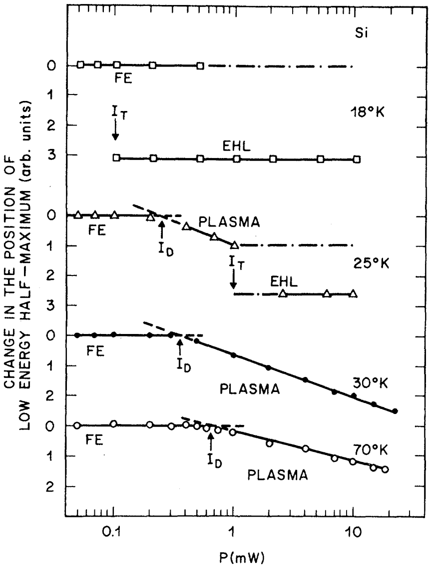

FIG. 2. Change in the position of low-energy half-maximum as a function of average incident power for various T. Upright arrows indicate I_D; inverted arrows indicate I_T. Points are experimental; dashed curves are extrapolations. Overlap with EHL prevents measurement of plasma curve (25 K) and exciton curve (18 K) at high intensities; expected behavior is shown by double-dashed lines.

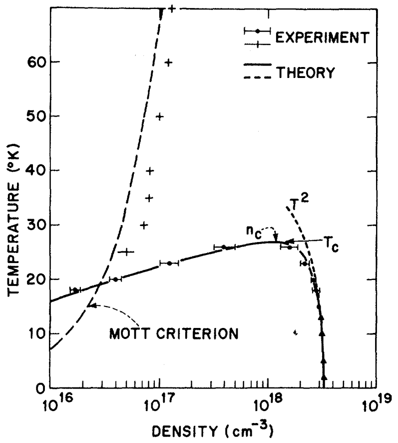

FIG. 3. Phase diagram of Si. Solid dots and crosses are our experimental data for coexistence curve and Mott dissociation, respectively. Triangles are from Ref. 6. Scaled phase diagram from Ref. 8 is shown by the solid curve. T^2 dependence of EHL density and the Mott criterion in the classical regime are shown by dashed curves.

the spectrum as a function of I for various T. The downward arrows in Fig. 2 indicate the threshold intensity I_T for EHL condensation. At 18 K the EHL condenses from FE for $I > I_T$. Since the line shapes of FE and EHL are independent of I, the half-maximum positions are constant and lie on two horizontal lines, one for each phase. At 25 K the FE line starts to broaden prior to the appearance of EHL. In Fig. 2 the half-maximum positions lie on a straight line which intersects the FE position at some point I_D indicated by the upward arrow. The sharp change occurring at I_D is attributed to onset of Mott dissociation of excitons into plasma. At 25 K the EHL appears at $I_T > I_D$ so that phase separation occurs between a plasma (P) and EHL, in contrast to the behavior at 18 K. The behavior for $T > T_c$ (T_c, the critical temperature for EHL formation, is 27 K as shown below) is also shown in Fig. 2 for $T = 30$ and 70 K. Since the plasma density increases with increasing I, the half-maximum position continues to shift toward lower energy. The width of the transition region, as well as the value of I_D, increases with increasing T.

The observed line shapes agree well with the theoretical FE line shape at low I and with the theoretical plasma line shape at high I as shown in Fig. 1(c).[8] Thus, the data presented here show that excitons dissociate into plasma with increasing I. This transition from FE to plasma is not abrupt, nor is it expected to be such at finite T.[1] We have determined I_D as a function of T from plots such as those in Fig. 2. Later in this Letter $I_D(T)$ will be converted into a density $n_D(T)$ and compared with the Mott criterion on $n_M(T)$ for exciton dissociation.

We now direct our attention to the determination of the phase diagram for EHL condensation in Si. The liquid side of the coexistence curve is obtained by determining the EHL density from its luminescence spectrum at a given T. The liquid spectrum is obtained by subtracting the spectrum at I just below I_T from the spectrum at I just above I_T where both phases coexist. This procedure is similar to that used by Thomas, Rice, and Hensel[9] For Ge. The liquid linewidth is used to obtain the corresponding liquid density from theoretical curves (half-width versus density) calculated by Hammond, McGill, and Mayer.[6] The resulting liquid side of the coexistence curve is shown in Fig. 3, where one finds that the liquid density varies as T^2 for low T with $n_0 = 3.3 \times 10^{18}$ cm^{-3} at $T = 0$.[6] We determine the critical temperture from the fact that two peaks in the spectra [see curve 3, Fig. 1(b)] are observed only for $T \leq 26$°K and that the behavior at 28°K is similar to that at higher T. Thus, $T_c = 27 \pm 1$ K.

A liquid-gas coexistence curve for Si has been calculated by Reinecke and Ying.[10] Their values for n_0 and T_c do not agree with our measurements. However, if we scale their calculated curve to our measured T_c and n_0, we get the solid curve shown in Fig. 3. We see that the agreement is quite good. From the fit we estimate that the critical density is $n_c \simeq 1.1 \times 10^{18}$ cm^{-3}. Combescot[11] calculated $T_c \simeq 28$°K and $n_c = 0.8 \times 10^{18}$ cm^{-3} whereas Vashishta, Das, and Singwi[12] calculated $T_c \simeq 20.8$°K and $n_c \simeq 1.2 \times 10^{18}$ cm^{-3}, and Reinecke and Ying[10] calculated $T_c \simeq 21.6$°K and $n_c \simeq 1.04 \times 10^{18}$ cm^{-3}.

The calculated coexistence curve extends to the gas side and predicts (when scaled to our T_c and n_0) that the gas density in equilibrium with EHL at 23°K is 1.25×10^{17} cm^{-3}. Thus, we take the measured I_T (0.62 mW) at 23°K to correspond to a density 1.25×10^{17} cm^{-3}. The experimental points on the gas side of the coexistence curve (see Fig. 3) are determined from measured I_T by using this conversion factor and assuming that density varies linearly with intensity.[13] Using the same conversion factor we have converted I_D into n_D, the density at which the exciton-plasma Mott transition occurs at various T; these n_D are also plotted in Fig. 3. The Mott criterion[1] predicts $n_M(T) = kT/16\pi E_x a^3 = (1.5 \times 10^{15})T$ cm^{-3} in the classical[14] regime and a limiting value of $n_M = 1.3 \times 10^{14}$ cm^{-3} at $T = 0$. Here E_x is the exciton binding energy (14.7 meV) and a ($= 44$ Å) is the exciton Bohr radius. We see in Fig. 3 that n_M is in excellent agreement with n_D.

We now discuss the possibility of a second peak[2] in the phase diagram due to the Mott transition. One would then expect a phase separation between FE and low-density plasma (LDP) for certain values of I and T. We have not observed any spectra with two distinct peaks indicative of FE-LDP phase separation. From Fig. 1 it seems possible for FE and LDP spectral peaks to be so close that their overlap results in a structureless band (e.g., curve 2, Fig. 1). In this case, subtraction of FE line shape from this band should yield a LDP spectrum whose shape is independent of I. We find that even for a 50% change in I near I_D the subtracted spectra differ from each other on the low-energy side. Yet the possibility of a narrow second peak in the phase diagram cannot be completely ruled out because of inhomogeneities resulting from surface excitation.

While the observed spectra at low and high intensities can be quantitatively understood in terms of FE and dense plasma or EHL, respectively, our understanding of the interesting intermediate region ($I \gtrsim I_D$) is inadequate at present. Neither the observed spectra, nor those obtained after subtracting the FE line, agree with the plasma line shape calculated in the standard manner.[6] Thomas *et al.*[15] have been able to fit their spectra in Ge close to I_D by adding the spectrum expected for excitonic molecule to the free-exciton spectrum. However, such a fit involves two fixed line shapes and is not expected to explain the continuous shift of the low-energy half-maximum for $I > I_D$ reported here in Fig. 2. In order to understand the intermediate region, one must know how to calculate the line shape of plasma with density between 10^{16} and 10^{17} cm^{-3}. No such calculation is available at present. It is clear that more theoretical and experimental work aimed at understanding the behavior in the FE-plasma transition region would be interesting and fruitful.

In conclusion, we have observed for the first time the dissociation of excitons into plasma predicted by Mott. The density at which the Mott transition occurs and its temperature variation are in good agreement with the Mott criterion. We have also obtained a phase diagram for electron-hole-liquid condensation in Si.

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[7] The choice of plotting the half-maximum is arbitrary. Plots similar to those shown in Fig. 2 are obtained for the change in positions of quarter- (three-quarter-) maximum except that I_D and n_D shift to lower (higher) values.

[8] The calculated plasma curve is shifted along the energy axis to obtain the best fit.

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[13] We have no experimental evidence to support this assumption. However, we find that the exciton luminescence intensity varies linearly with I. If the volume occupied by the excitations at 70-nsec delay varies sufficiently slowly with I and T, deviations from linear behavior may introduce effects within the experimental error bars. Note also that I_D and I_T are at least 30 times smaller than the laser power used in Ref. 5. Hence the initial density and expansion rate are considerably smaller. At 70-nsec delay we find that complete thermalization takes place.

[14] This expression differs from the expressions given in Refs. 3 and 9 because both electrons and holes contribute to screening.

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Kondo Lattice: Real-Space Renormalization-Group Approach

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A one-dimensional analog of the Kondo lattice is studied by a renormalization-group technique. Previous mean-field results are shown to be reasonable: The system undergoes a second-order crossover transition from an antiferromagnetic state to a Kondo-like state at zero temperature as the spin–conduction-electron coupling is increased. Estimates are given for critical exponents and the behavior of correlation functions near the transition.

$CeAl_2$, $CeAl_3$, and many rare-earth compounds[1] behave anomalously at low temperature—either they do not order magnetically or they have a very low transition temperature. Several authors[1-3] attribute this to a Kondo effect. While the Kondo problem is now solved[4] for a single magnetic impurity, the Kondo-lattice problem which involves one impurity per cell remains an open question. In order to investigate the general properties of this kind of system, Doniach[5] has previously introduced a simple one-dimensional (1D) analog Hamiltonian, the "Kondo necklace":

$$H = J\sum_i \vec{S}_i\cdot\vec{\tau}_i + W\sum_i(\tau_i^x\tau_{i+1}^x + \tau_i^y\tau_{i+1}^y), \qquad (1)$$

where J is positive and where $\vec{S}_i$ and $\vec{\tau}_i$ are two independent sets of Pauli operators $[\tau_i^x = \left(\begin{smallmatrix}0&1\\1&0\end{smallmatrix}\right)$, etc.].

In this Hamiltonian the 1D electron gas has been replaced[5,8] by a set of pseudo spins $\frac{1}{2}\vec{\tau}_i$ regularly spaced on an infinite linear lattice.

The general qualitative behavior of this model may be seen from the weak- and strong-coupling limits: $J/W = 0$ and ∞. For $J = 0$, it reduces to the X-Y quantum-spin chain,[7] which is equivalent to a 1D spinless Fermi gas with one electron per atom.[6-8] For small J/W, we expect that the S spins couple antiferromagnetically via the τ spins, leading to a ground state of broken symmetry exhibiting characteristic spin-wave-like excitations. On the other hand, when $J/W = \infty$, the system reduces to a set of noninteracting singlet-triplet cells leading to a singlet ground state for (1) well separated by a gap of $4J$ from the first excited states. So, we expect a transition from a magnetic behavior to a Kondo-like behavior by increasing the ratio J/W. Within a mean-field approximation,[5,9] this transition occurs at $(J/W)_c = 1$. But it is well known that fluctuation effects, in one dimension, strongly affect the mean-field results. The purpose of this Letter is to present a renormalization-group approach to the problem. Our results confirm the general qualitative features of the model previously suggested.[5,9] We find a critical value $(J/W)_c \cong 0.4$ at which there is a second-order transition at zero temperature in which the system crosses over from antiferromagnetic to Kondo-like behavior. The critical indices for this fixed point are estimated and found to be similar (but not identical) to those for an Ising chain in a transverse field. The approach appears to be generalizable to other quantum spin systems.