Phase transition and Coulomb correlation of impurity-band electrons at the surface of a semiconductor at nonzero temperatures

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The Coulomb interaction of electrons localized on monoenergetic capture centers located on an interface is examined for nonzero temperatures. The free energy of an electron system with a Coulomb interaction is calculated. The result is used to derive the Fermi level μ, the correlation radius r_c, and the heat capacity as functions of the temperature. When the impurity band is filled only slightly, a phase transition occurs as the temperature is changed; at a low temperature, a phase transition occurs as the band filling changes. At the transition, r_c changes abruptly. There is a singularity in the dependence of μ on the temperature and the concentration at the transition. It has been predicted elsewhere that μ would be a nonmonotonic function of the density of charged centers at zero temperature because of the correlation in the positions of these centers. This nonmonotonic behavior gradually disappears as the temperature is raised. The state density in the impurity band is calculated.

1. INTRODUCTION

It is becoming progressively more obvious that at low temperatures the Coulomb interaction can play an important role in shaping the energy spectrum of an impurity band at the surface of a semiconductor.

A correlation caused in the positions of charged centers by the Coulomb interaction gives rise to a Coulomb gap in the energy spectrum of localized states.[1,2] In the limiting case of complete correlation, the system could become a Wigner crystal of charged centers.

We have previously[3] examined the consequences of a correlation among charged donor states at an insulator-semiconductor interface at zero temperature. We showed that the Coulomb interaction causes the position of the Fermi level to be a nonmonotonic function of the degree of filling, with the further consequence that there is a negative effective surface-state density. A Coulomb gap arises in the state density.

Similar results have emerged from a numerical simulation by Bello *et al.*[4] The agreement of the results derived in Refs. 3 and 4 lends support to the approximations adopted in Ref. 3 and to the approach of using the cell model to describe the many-particle Coulomb interaction. The reason for this circumstance is the small-scale nature of the two-dimensional fluctuational potential.[5,6]

We wish to emphasize that the approach taken in Ref. 3, of partitioning the surface into cells and averaging the potential over each cell, makes it possible to describe the correlation of charged centers in a common way, by beginning from a complete correlation (a Wigner crystal), with an impurity band filled only slightly, and proceeding all the way to a complete absence of correlation (a random distribution of the charged centers over the surface), with a highly filled band.

In this paper we explain how a nonzero temperature affects the correlation of charged centers at a surface. A nonzero temperature is one more factor tending to cause disorder in the system. We will thus examine the joint effects of a random distribution of impurity centers and a nonzero temperature on the correlation.

Let us consider the interface between a semiconductor and an insulator. We assume that there are monoenergetic capture centers here with an energy $-E_0$ reckoned from the bottom of the conduction band of the semiconductor, and with a concentration σ. If a center has not been filled, it is neutral; when a center captures an electron, the local state becomes negatively charged.

We assume that the capture centers are distributed in a random fashion over the surface and have a Poisson distribution

$$P_m(r) = \frac{(\pi r^2 \sigma)^m}{m!} e^{-\pi r^2 \sigma},$$

which gives the probability for finding m centers in a circle of radius r if the average concentration of states is σ. We showed in Ref. 3 that the correlation can be characterized by some correlation radius r_c. We can find r_c as a function of the filling of the capture centers at zero temperature by using simply the fact that the charges at the surface repel each other. This repulsion gives rise to an ordering of the charges, and in principle the preferred structure at the surface is a triangular lattice. A random distribution of local centers over the surface disrupts the order of this lattice, because some lattice sites may not have a center, and an electron will be forced to stay near this site, at the nearest local center. As the number of vacant positions decreases (as the degree of filling is increased), the ordering decreases. Nevertheless, the interface can be partitioned into cells of radius r_c such that each cell contains only a single charged center if it has at least one center. The concentration of charged centers, σ_-, can then be written[3]

$$\sigma_- = \frac{1}{\pi r_c^2} \sum_{m=1}^{\infty} P_m(r_c) = \frac{1}{\pi r_c^2} [1 - \exp(-\pi r_c^2 \sigma)].$$

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The exponential function on the right side of (1) gives the relative number of cells which have no centers. Denoting by $\eta = \sigma_-/\sigma$ the extent to which the impurity band is filled and by $\xi = \pi r_c^2 \sigma$ the average number of centers per circle of radius r_c, we can convert the expression for σ_- to the form

$$\eta = (1 - e^{-\xi})/\xi, \qquad (1)$$

from which we see that the order parameter is the quantity

$$\theta = \eta \xi = 1 - e^{-\xi}.$$

For a Wigner crystal, which would exist at a slight filling ($\eta \ll 1$), we would have $\theta = 1$, while in the absence of correlations ($\xi = 0$) we would have $\theta = 0$.

2. FREE ENERGY OF THE SYSTEM

We now consider a system of surface centers at a nonzero temperature. We assume that on a surface of area S there are $B = \sigma S$ centers at which there are N electrons. We assume that the first Bohr radius of a local state, a_B, is much smaller than the average distance between centers. We can then ignore the overlap of the wave functions of the different centers and correspondingly treat the electrons at the surface as constituting a lattice gas without a kinetic energy. In this case the partition function Z is determined exclusively by its configurational part, which can be written in the present case as

$$Z = \frac{1}{N!} \sum_{\substack{i=1 \\ r_1}}^{B} \cdots \sum_{\substack{i=1 \\ r_N}}^{B} \exp\left[- \sum_{l,k=1}^{N} U(r_{k,l})/2kT \right], \qquad (2)$$

where $U(r_{k,l})$ is the energy of the Coulomb interaction between the k th and l th electrons; a neutralizing background has been taken into account. The summation is over all the coordinates of the B centers at the surface for all N electrons.

Although expression (2) for Z is rigorous, we cannot carry out exact calculations from it. We will accordingly try to simplify it. We first note that for a two-dimensional distribution of charges the potential fluctuations are due primarily to the small-scale potential.[5,6] In determining the potential in a cell, we can thus average the random potential over the positions of the charges outside the cell of radius r_c, as in Ref. 3. As a result we find the potential energy of an electron in a cell:

$$\left\langle \sum_{k=1}^{N} {}' U(r_{k,l}) \right\rangle = -\frac{4q^2 r_c \sigma_-}{\varepsilon^*} E\left(\frac{r}{r_c}\right) = \varepsilon(r), \qquad (3)$$

where $\varepsilon^* = (\varepsilon_d + \varepsilon_s)/2$ is half the sum of the dielectric constants of the insulator and the semiconductor, σ_- is the average surface density of electrons, $E(y)$ is a complete elliptic integral of the second kind, r is the distance from the center of the cell, and the prime on the summation sign means that the summation is over all the electrons except that in the cell under consideration. At this point we replace $E(y)$ by the approximate expression $E(y) \approx \frac{1}{2}\pi(1 - \frac{1}{4}y^2)$. Furthermore, in partition function (2) we ignore configurations which allow for the possibility of two or more electrons in a single

cell. This step is justified by the appearance of an additional positive energy because of the Coulomb repulsion among the electrons. The model developed below is in the spirit of the hole theory of the liquid state.[7,8] There is a difference from that theory, in that here we are introducing a Poisson distribution of impurity centers, so that there are unfilled cells, not only because of thermal excitation but also because there are cells without impurity centers.

We now group all the cells. Each cell in each group has m local states with coordinates $\{r_i\}$, where r_i is the distance from the ith state to the center of the cell, with i running from 1 to m. We denote the number of cells in a group by G_{ml}, and we denote the number of electrons by N_{ml}, where the vector l is determined by the set of coordinates $\{r_i\}$. It is then a simple matter to rewrite partition function (2) as

$$Z = \prod_{m,1} \frac{G_{ml}!}{N_{ml}!(G_{ml} - N_{ml})!} \left(\sum_{i=1}^{m} \exp\left\{ -\frac{\varepsilon(r_i)}{2kT} \right\} \right)^{N_{ml}}, \qquad (4)$$

where $\varepsilon(r_i)$ is given by (3). The free energy of the system is then

$$F = -kT \ln Z = kT \sum_{ml} G_{ml} \left\{ n_{ml} \ln n_{ml} \right. $$
$$\left. + (1 - n_{ml}) \ln(1 - n_{ml}) - n_{ml} \ln \sum_{i=1}^{m} \exp\left[-\frac{\varepsilon(r_i)}{2kT} \right] \right\}, \qquad (5)$$

where $n_{ml} = N_{ml}/G_{ml}$ is the filling function of a cell with configuration (m,l) The number of cells in the group G_{ml} is

$$G_{ml} = \frac{S}{\pi r_c^2} P_m(r_c) m \left(1 - \frac{r_1^2}{r_c^2} \right)^{m-1} d\left(\frac{r_1^2}{r_c^2} \right) \prod_{i=2}^{m} \frac{dr_i^2}{r_c^2 - r_1^2}, \qquad (6)$$

where $SP_m(r_c)/\pi r_c^2$ is the number of cells on the surface with m local states. The factor $m(1 - r_1^2/r_c^2)^{m-1} d(r_1^2/r_c^2)$ is the probability for the appearance of a state in the ring between r_1 and $r_1 + dr_1$ under the condition that there is no state in the circle of radius r_1. The factor $dr_i^2/(r_c^2 - r_1^2)$ is the probability for the appearance of one of the $m - 1$ remaining states in the ring between r_i and $r_i + dr_i$. Introducing $V_\sigma = 2q^2(\pi\sigma)^{1/2}\varepsilon^*$ and $x = r^2/r_c^2$, we can rewrite $\varepsilon(r)$ as $\varepsilon(r) = V_\sigma \eta \xi^{1/2}(1 - x/4)$ and determine the normalized reciprocal temperature $\beta = V_\sigma/kT$. As a result, free energy (5) becomes

$$F = kT\sigma S \left\{ -\frac{1}{2} \beta \eta^2 \xi^{1/2} \right.$$
$$+ [n_{ml} \ln n_{ml} + (1 - n_{ml}) \ln(1 - n_{ml}) \qquad (7)$$
$$\left. - n_{ml} \ln \left(\sum_{i=1}^{m} \exp(-\beta \eta \xi^{1/2} x_i/8) \right) \right]_{av} \right\}.$$

The subscript "av" on the square bracket denotes an average over all the configurations (m,l):

$$(A_{ml})_{av} = \sum_{m=1}^{\infty} e^{-\xi} \frac{\xi^{m-1}}{(m-1)!} \int_0^1 (1 - x_1)^{m-2} dx_1 \prod_{i=2}^{m} \int_{x_1}^{1} dx_i A_{ml}. \qquad (8)$$

The extent to which the states are filled, η, is given by

$$\eta = (n_{ml})_{\mathrm{cp}}. \tag{9}$$

To determine the Fermi level μ, we introduce the thermodynamic potential Ω:

$$\Omega = F - \mu N. \tag{10}$$

3. EQUATIONS OF STATE

In the preceding section we derived expressions for the thermodynamic potentials and the extent to which the states are filled, as functions of the temperature, the cell filling functions n_{ml}, and the correlation radius ξ. We find the Fermi level of the system by minimizing Ω with respect to n_{ml}:

$$\delta\Omega/\delta n_{ml} = 0. \tag{11}$$

The result is

$$\frac{\mu}{kT} = -\beta\eta\xi^{1/2} + \ln\frac{n_{ml}}{1-n_{ml}}$$
$$- \ln\sum_{i=1}^{m}\exp\left(\frac{-\beta\eta\xi^{1/2}x_i}{8}\right) + \frac{1}{8}\beta\xi^{1/2}n_{ml}\langle x_i\rangle, \tag{12}$$

where

$$\langle x_i\rangle = \sum_{i=1}^{m}x_i\exp\left(\frac{-\beta\eta\xi^{1/2}x_i}{8}\right)\bigg/\sum_{i=1}^{m}\exp\left(\frac{-\beta\eta\xi^{1/2}x_i}{8}\right).$$

From Eq. (12) we find an expression for the cell filling function n_{ml}:

$$n_{ml} = \left\{1 + \left[\sum_{i=1}^{m}\exp(-\beta\eta\xi^{1/2}x_i/8)\right]^{-1}\right.$$
$$\left.\times\exp[-\mu/kT - \beta\eta\xi^{1/2}(1-n_{ml}\langle x_i\rangle/8\eta)]\right\}^{-1}. \tag{13}$$

We see from (13) that the cell filling function is a Fermi function which allows any of the states in the cell to be filled with a corresponding statistical weight.

Expression (9) with (13) is an integral equation which determines the relationship between the Fermi level and the degree of filling η at a given temperature and at a given correlation radius ξ.

The correlation radius is an internal parameter of the system and can be found by minimizing F for a given value of η:

$$\partial F/\partial\xi\big|_{\eta} = 0. \tag{14}$$

The optimization with respect to ξ allows for possible changes in the arrangement of the charged centers.

From (14) we find an additional equation which relates ξ, n_{ml}, η, and β:

$$\frac{\beta\eta^2}{4\xi^{1/2}}\left[1 - \frac{(n_{ml}\langle x_i\rangle)_{\mathrm{av}}}{4\eta}\right] = \left[\left(1 - \frac{m-1}{\xi}\right)\ln(1-n_{m,1})\right]_{\mathrm{av}}. \tag{15}$$

To solve system of equations (9), (13), (15), we begin by analyzing these equations.

To show that the cell filling function n_{ml} is a negligibly weak function of the particular configuration (m,l), we introduce the function $n_{ml}(x_i)$, which gives the filling of the ith state in the cell. The filling of the ith state can be expressed in terms of the filling of the first state:

$$n_{ml}(x_i) = n_{ml}(x_1)\exp[-\beta\eta\xi^{1/2}(x_i-x_1)/8], \tag{16}$$

The cell filling function is equal to the sum of the filling functions of each of the states in the cell:

$$n_{ml} = \sum_{i=1}^{m}n_{ml}(x_i). \tag{17}$$

Let us examine the filling n_{ml} in the two limiting cases of low and high temperatures. It follows from expression (16) that at a low temperature $(\beta\eta\xi^{1/2}/8\gg1)$ the filling of the ith centers $(i = 2,...,m)$ is negligible in comparison with the filling of the first. Furthermore, since (as we will see below) the Fermi level is quite high in units of kT in comparison with the energy of the filled states, the filling of a cell depends exponentially weakly on the position of the first state also.

At a high temperature the correlation radius tends toward zero, as we will see below, and the probability for the appearance of two or more states in such cells also tends toward zero. Consequently, again at high temperatures we need consider the filling of only the first state, and we may assume that the filling of a cell is independent of its position.

We thus assume that the filling of a cell is independent of (m,l) over the entire temperature range, and we replace n_{ml} by the average cell filling n. This is the approximation of a self-consistent field, in which all the cells which contain at least one state are assumed to be identical and are assumed to be in an identical average self-consistent potential. In expression (7) for the free energy we thus replace n_{ml} by n, and in taking the average in the last term we assume that the filling of $m - 1$ states is small in comparison with that of the first state. As a result we find

$$F = \sigma S V_\sigma\left\{-\frac{1}{2}\eta^2\xi^{1/2} + \frac{\eta}{\beta}\left[\ln n + \left(\frac{1}{n} - 1\right)\ln(1-n)\right]\right.$$
$$\left.- \frac{8\xi^{1/2}}{\beta^2}\left[1 - \frac{\xi}{1-e^{-\xi}}\frac{e^{-\xi} - e^{-\beta\eta\xi^{1/2}/8}}{\beta\eta\xi^{1/2}/8 - \xi}\right]\right\}. \tag{18}$$

The last term in the curly brackets is correct only at low temperatures. Although this term is small, it proves important in calculating the low-temperature specific heat; it should be ignored at high temperatures.

In this self-consistent field approximation, expression (9) becomes

$$\eta = n(1-e^{-\xi})/\xi, \tag{19}$$

and the order parameter θ is

$$\theta = n(1-e^{-\xi}). \tag{20}$$

The quantity in parentheses in (19) and (20) is the relative number of cells which contain at least one state, and n is the probability for the filling of these cells. Comparing (1) and (19) we see that an additional randomization occurs at a nonzero temperature in the system because of the thermal excitation of cells $(n < 1)$.

In the self-consistent field approximation, Eq. (15) reduces to an equation which describes the relationship between the temperature and ξ at various values of η:
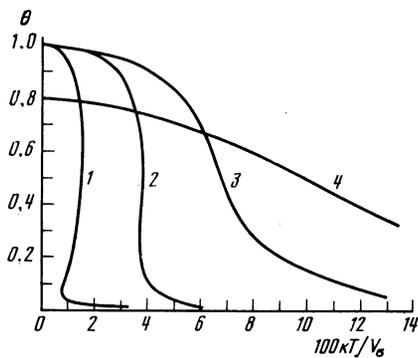
334   Sov. Phys. JETP **59** (2), February 1984

E. V. Chenskiĭ and Yu. Ya. Tkach   334

FIG. 1. Temperature dependence of the order parameter θ for various degrees of filling η. 1—$\eta = 0.02$; 2—0.1; 3—0.2; 4—0.5.

$$\beta = -\frac{4[1-(1+\xi)e^{-\xi}]}{\eta^2\xi^{3/2}}\ln\left(1-\frac{\eta\xi}{1-e^{-\xi}}\right). \qquad (21)$$

From (19) and (21) we have

$$n \to 1 \quad \text{as} \quad T \to 0;$$

$$n \to \eta, \quad \xi \to 0 \quad \text{as} \quad T \to \infty,$$

from which we see that in the limit $T \to 0$ expression (19) reduces to (1), which was derived for a zero temperature.

Working from (21), we can calculate the temperature dependence of the order paramater θ for various fillings η (Fig. 1) and also the η dependence of θ at various temperatures (Fig. 2). We see from Fig. 1 that at $\eta > 0.12$ the order parameter θ falls off monotonically with increasing temperature. At $\eta < 0.12$ we see an S-shaped $\theta(T)$ dependence, which corresponds to a first-order phase transition from an ordered state to a disordered state. At $\eta = 0.12$ the difference between the ordered and disordered phases disappears; i.e., this is a critical value of η. As η is reduced from 0.12, the jump in the order parameter at the phase transition, $\Delta\theta$, increases, and in the limit $\eta \to 0$ we find $\Delta\theta \to 1$.

It follows from Fig. 2 that at a low temperature, $T < T_{cr}$ ($kT_{cr} = V_\sigma/25$), there is an S-shaped dependence of θ on the degree of filling; a first-order phase transition occurs as the filling is varied, and the order parameter changes abruptly. At $T > T_{cr}$ the order parameter changes continuously with η.
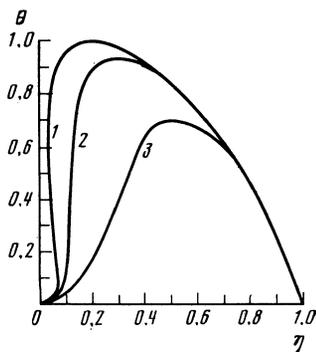

FIG. 2. The order parameter θ vs the degree of filling η for various temperatures. 1—$kT = v_\sigma/100$; 2—$KT = V_\sigma/25$; 3—$kT = V_\sigma/5$.

What is the physical meaning of these phase transitions? The ordering factor in this system is the mutual Coulomb repulsion of the charges; the interaction intensifies with increasing concentration of charged centers. With increasing filling of the impurity band, on the other hand, the system becomes more random because of the random positions of the local centers on the surface.

The temperature is also a randomizing factor, having its greatest effect at a slight filling. The presence of these three competing factors thus gives rise to phase transitions as either the temperature or the filling is varied. The joint effects of the temperature and the filling on the randomization of the system lead to a critical point on the (η, T) phase diagram, where the difference between the ordered and disordered phases disappears (Fig. 4).

The transition temperature at $\eta \ll 1$ can be determined by comparing the free energies of the ordered and disordered states. The free energy for the ordered state is determined primarily by the interaction energy,

$$F \approx -{}^{1}/_{2}V_\sigma\sigma\eta^{4/3},$$

while that for the disordered state is determined by the entropy term,

$$F \approx kT\sigma\eta\ln\eta.$$

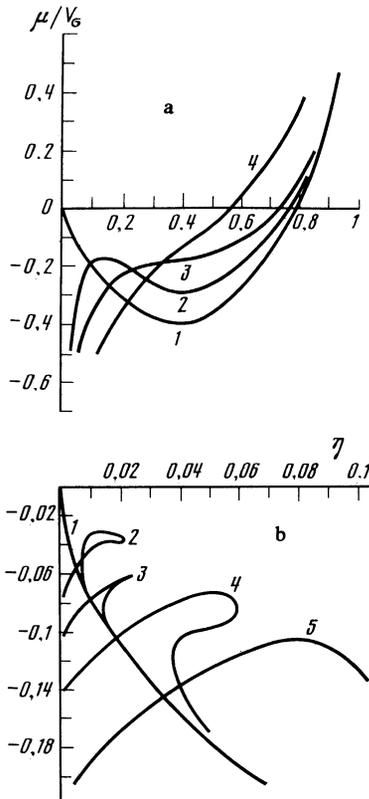Equating these expressions, we find the transition temperature T_c:

FIG. 3. Position of the Fermi level, μ, vs the degree of filling at various temperatures. a: 1—$T = 0$; 2—$kT = V_\sigma/20$; 3—$kT = V_\sigma/5$; 4—$kT = V_\sigma$. b: 1—$T = 0$; 2—$kT = V_\sigma/100$; 3—$kT = V_\sigma/55$; 4—$kT = V_\sigma/40$; 5—$kT = V_\sigma 25$.
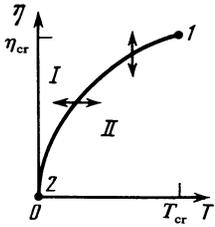
E. V. Chenskiĭ and Yu. Ya. Tkach   335

FIG. 4. The (η,T) phase diagram. I: Ordered state (Wigner glass). II: Disordered state (gas). 1—Critical point $(\eta_{cr} = 0.12,\ kT_{cr} = V_\sigma/25)$; 2—isolated singularity corresponding to a Wigner crystal; horizontal arrow—phase transition as the temperature is varied; vertical arrow—phase transition as the filling is varied.

$$kT_c \approx -V_\sigma \eta^{1/2}/2 \ln \eta.$$

As was shown in Ref. 4, there should be no long-range order in the electron structure in a random lattice of localized states, even at a slight filling. The order parameter θ which we have derived is a measure of the short-range order in the system, and we are actually dealing with a Wigner glass-gas transition. Long-range order arises only asymptotically in the limit $\eta\to0$, $T\to0$ in a Wigner crystal; i.e., on the (η,T) phase diagram the Wigner crystal is an isolated singularity at the origin (Fig. 4).

Now working from the equation $\mu = \partial F/\partial\eta$ and expression (18) for F (this expression was derived in the self-consistent-field approximation), we find the position of the Fermi level as a function of the filling and the temperature:

$$\mu = -V_\sigma \eta \xi^{1/2}\left[1 - \frac{\eta\xi}{4[1-(1+\xi)e^{-\xi}]}\right] + kT \ln\frac{\eta\xi}{1-e^{-\xi}}. \quad (22)$$

Let us consider some limiting cases. At a low temperature we find from system (21), (22)

$$\mu = -{}^3/_4 V_\sigma \eta^{1/2}, \quad \eta\ll1;$$
$$\mu = V_\sigma/2[2(1-\eta)]^{1/2}, \quad 1-\eta\ll1. \quad (23)$$

At a high temperature $(kT\!\gg\!V_\sigma\,\eta\xi^{1/2})$ we find from (21), (22) the ordinary behavior of the Fermi level, which we would
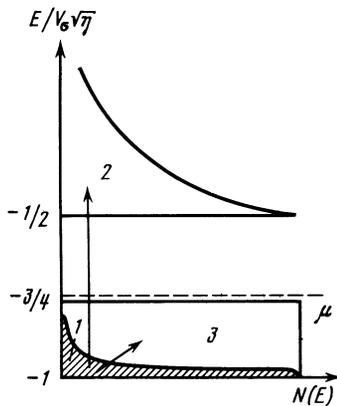

FIG. 5. Density of states and density of excitations in a system with a Coulomb gap for a slight filling and a low temperature. 1 (hatched region)—Filled states; 2—unfilled states; 3—gap-free density of excitations; vertical arrow—excitations across the Coulomb gap; inclined arrow—excitations without a gap.

have for a monoenergetic level in the absence of a Coulomb interaction:

$$\mu = kT \ln\frac{\eta}{1-\eta}. \quad (24)$$

System (21), (22) thus describes the known limiting transitions for the low-temperature case, for both slight and pronounced filling,[3,4] and also for the high-temperature case.

Figure 3 shows the filling dependence of the position of the Fermi level at various temperatures. Figure 3a shows $\mu(\eta)$ at $T = 0$ and at $T > T_{cr}$. At low temperatures, the function $\mu(\eta)$ is nonmonotonic. At $kT = V_\sigma/5$ it becomes monotonic, and at $kT\!\gg\!V_\sigma$ we find the usual $\mu(\eta)$ dependence [see (24)]. Figure 3b shows $\mu(\eta)$ for $T < T_{cr}$ in the region $\eta < \eta_{cr}$. Structural features appear on the $\mu(\eta)$ curve because of the phase transition as the filling is varied.

If the impurity band of interest is at a semiconductor-insulator interface in a metal-insulator-semiconductor structure, then the phase transitions which occur as the filling or the temperature is changed will be seen as an unbounded increase in the capacitance of the metal-insulator semiconductor structure and as abrupt changes in the charge in the impurity band and at the gate.

We can now calculate the heat capacity of the system, C, which is defined by

$$C = -T(d^2F/dT^2). \quad (25)$$

Substituting Eq. (21), which relates T and ξ, along with F from (18) into (25), we find the heat capacity as a function of the temperature. The results of numerical calculations show that at $\eta > \eta_{cr}$ the heat capacity has a slight maximum; at $\eta = \eta_{cr}$, at the critical point, C diverges; and at $\eta < \eta_{cr}$, the behavior of $C(T)$ is the same as that at a first-order phase transition.

At low temperatures (far from the phase transition) and at $\eta\!\ll\!1$ the heat capacity is given by

$$C = \frac{16\sigma k^2 T}{V_\sigma \eta^{1/2}} + \frac{V_\sigma^2 \eta^3}{8kT^2}\exp\left\{-\frac{V_\sigma \eta^{1/2}}{4kT}\right\}. \quad (26)$$

It follows from this expression that there are two types of excitations: zero-gap excitations and excitations across a gap. The zero-gap excitations, which are described by the first term in (26), are related to transitions within a cell and dominate the heat capacity at low temperatures. The density of excitations in a cell was calculated in Ref. 3. It can also be found from (26), by comparing the first term there with the known expression for the heat capacity of a degenerate electron gas, which depends on the state density at the Fermi level.

As the temperature is raised, excitation across a gap becomes important and is responsible for structural changes (changes in the correlation radius); in particular, it determines the phase transition. The excitations across a gap depend exponentially on the temperature, so that the distance from the Fermi level to the position of the unfilled states can be determined from the argument of the exponential function. It follows from (26) that this distance is $V_\sigma\,\eta^{1/2}/4$ if the filling is slight and the temperature is low. As we saw earlier, the distance from the Fermi level to the level of the filled

states is also equal to $V_\sigma \, \eta^{1/2}/4$. It follows that the size of the Coulomb gap is $V_\sigma \, \eta^{1/2}/2$. Figure 5 is an energy diagram of the density of states and the density of excitations.

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