# Effect of quantum hopping on the Coulomb gap of localized electrons in disordered systems

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In this paper, we use the coherent potential approximation to study the role quantum hopping plays on the Coulomb gap of insulating strongly correlated d=2 and d=3 dimensional systems. We find that substantial increase in the density of states at the Coulomb gap occurs only when the ratio between the hopping integral t and the gap width B exceeds a critical value. We estimate that the hopping integral corresponding to the experimental condition  $n < n_c/3$  satisfies t/B < 0.05. For such values of t/B, quantum hopping brings about little change in the single particle density of states. The classical Coulomb gap therefore remains intact in both three and two dimensional systems, in contrast to a previous claim that the gap disappears for d=2 systems. The implication of these results on experiments on doped semiconductors is discussed.

#### I. INTRODUCTION

The presence of the long-range Coulomb interactions in disordered materials is believed<sup>1</sup> to diminish the single particle density of states close to the Fermi level,  $E_F$ . Efros and Shklovskii<sup>2</sup> have shown that at T=0, a soft gap of the form  $(E - E_F)^{d-1}$  results. A gap of this form has a profound effect on the transport properties of strongly localized systems. For example, the Mott variable-range-hopping law is transformed to a universal form independent of dimensionality<sup>2</sup>. Experimental observation of hopping conductivity of the form  $\ln \sigma(T) \sim T^{-\alpha}$  with  $\alpha$  close to 1/2 in doped semiconductors has been widely interpreted as a confirmation of the presence of a soft Coulomb gap. There are still, however, several outstanding problems with the soft Coulomb gap account.3 The major questions appear to be the following. (1) Is the single particle distribution sufficient to describe the low-energy excitations? (2) Does quantum tunneling wash-out the soft Coulomb gap? (3) How does the filling-in of the Coulomb gap at finite temperatures affect the temperature dependence of the conductivity?

In a previous paper,4 we showed that finitetemperature effects profoundly influence the form of the conductivity. Specifically, we showed that if the finitetemperature configuration of particles and holes is used to compute the conductance, activated rather than  $T^{-1/2}$ temperature dependence is observed. In the present study, we address the question of the role of quantum tunneling on the soft Coulomb gap. Recent theoretical studies have found that tunneling can counteract the tendency of long-range interactions to create a gap in the density of states. Vignale and co-workers concluded that a finite density of states is created at the Fermi level in d=2 for any nonzero hopping matrix element<sup>5</sup>. A less drastic effect was found for d = 3, namely, only a narrowing of the gap depending on the magnitude of

the hopping integral. The apparent disappearance of the Coulomb gap in 2d when quantum hopping is taken into account has been cited<sup>5,6</sup> as a possible explanation of experiments<sup>7</sup> on d=2 Si metal-oxide semiconductor field-effect transistors (MOSFET's) that fail to observe the Efros-Shklovskii (ES) form of the conductivity under conditions that favor the formation of a Coulomb gap. Other indications of the importance of quantum hopping include the observation of large conductance fluctuations in materials exhibiting hopping conduction,<sup>8</sup> a phenomenon usually associated with quantum coherence.

In view of the relevance of quantum tunneling to experimental observations, we reexamine its role on the emergence of a soft Coulomb gap in disordered systems. The specific question we address is, how large should the hopping integral be before any discernible enhancement in the number of states at the Fermi level is obtained? Following Vignale and co-workers<sup>5</sup> the starting point for our analysis is the standard tight-binding model of an impurity band in a doped semiconductor. We then compute the density of states using the coherent potential approximation (CPA). We find that for realistic values of the hopping integral, t, the Coulomb gap remains intact for both d equal to two and three dimensional systems. For example, we find that the increase in the density of states is less than 5% when t/B = 0.05, where B is the gap width. This value corresponds to the maximum hopping integral allowed at an impurity density of 1/3 the critical concentration at which the system becomes a metal. Because most experiments are performed at impurity concentrations less than this value, we argue that quantum hopping should not play a significant role in enhancing the density of states at the Fermi level. This paper is organized as follows. The theoretical formalism is introduced in Sec. II. Section III contains an analytical solution to the CPA equations in the vicinity of the Fermi energy. Numerical solutions as well as a discussion are presented in Secs. IV and V, respectively.

#### II. FORMALISM

We start our analysis with the effective tight-binding Hamiltonian

$$H = \sum_{i} \epsilon_{i} n_{i} + \sum_{i \neq j} t_{ij} c_{i}^{\dagger} c_{j} , \qquad (1)$$

where  $c_i^{\dagger}$  and  $c_i$  are creation and annihilation operators of spinless electrons on site i, respectively. The first part of the Hamiltonian is the Hartree site energy in the ground state of the interacting classical Hamiltonian. The second part describes quantum hopping.  $t_{ij}$  is the hopping integral between site i and j and has the translational symmetry of a periodic lattice. The exact form of  $t_{ij}$  will not be specified in the following calculation. Site energies  $\epsilon_i$  in the Coulomb gap problem are highly correlated among neighboring sites. However, for the questions we are interested in, we can assume that  $\epsilon_i$  is independently distributed with probability  $P(\epsilon)$ . We assume then that  $P(\epsilon)$  is given by the ES distribution<sup>2</sup>

$$P(\epsilon) = c\epsilon^{d-1} \tag{2}$$

for  $|\epsilon| < B$  and  $P(\epsilon) = 0$  otherwise, where  $c = \frac{d}{2B^d}$ .

The above Hamiltonian is appropriate only when the quantum hopping term is small relative to the width of the site energy distribution such that all states are strongly localized. By not including spins, we also neglect possible exchange couplings between electrons. Such couplings are essential to correctly describe the magnetic properties and may even be responsible for a crossover from variable range hopping form of conductance to an activated form, upon reducing temperature.<sup>9</sup>

The density of states in this system can be calculated through the CPA technique. The effective single particle Green function is<sup>10</sup> defined as

$$G(\omega) = \langle G_{ii}(\omega) \rangle = G_0[\omega - \Sigma(\omega)] , \qquad (3)$$

where

$$G_0(\omega) = \left(\frac{1}{\omega - H_0}\right) ii = \int \frac{g(\omega)d\omega}{\omega - \epsilon}.$$
 (4)

 $g(\omega)$  and  $H_0$  are the density of states and the Hamiltonian in the absence of the site disorder, respectively.  $\Sigma(\omega)$  is the self-energy.  $\langle \cdots \rangle$  denotes the average over disorder configurations. The single particle density of states is the imaginary part of the Green function,

$$\rho(\omega) = -\frac{1}{\pi} \text{Im} G(\omega + i0)$$

The equation for the self-energy at the CPA level is given by  $^{10}$ 

$$\int \frac{P(\epsilon)d\epsilon}{1 - [\epsilon - \Sigma(\omega)]G(\omega)} = 1.$$
 (5)

We will find it more convenient to define a new quantity  $(\xi = \xi_1 + i\xi_2)$ ,

$$\xi(\omega) = \Sigma(\omega) + \frac{1}{G(\omega)} \tag{6}$$

and recast Eq. (5) as

$$G(\omega) = \int \frac{P(\omega)d\epsilon}{\xi(\omega) - \epsilon}.$$
 (7)

For the distribution given in Eq. (2), we can integrate Eq. (7) exactly to obtain

$$G(\omega) = c\xi \left[ -2B + \frac{\xi}{2} \ln \frac{(B+\xi_1)^2 + \xi_2^2}{(B-\xi_1)^2 + \xi_2^2} - i\xi \arctan \frac{B-\xi_1}{\xi_2} + i\xi \arctan \frac{B+\xi_1}{\xi_2} \right]$$
(8)

for d=3, and

$$G(\xi) = \frac{\xi}{B} \left[ -\frac{1}{2} \ln \frac{[(B-\xi_1)^2 + \xi_2^2][(B+\xi_1)^2 + \xi_2^2]}{[\xi_1^2 + \xi_2^2]^2} -2i \arctan \frac{\xi_1}{\xi_2} - i \arctan \frac{B-\xi_1}{\xi_2} + i \arctan \frac{B+\xi_1}{\xi_2} \right]$$
(9)

for d=2. To find the density of states, we must solve Eqs. (3), (4), and (6) simultaneously with Eq. (8) for d=3 and Eq. (9) for d=2. In general, numerical techniques have to be employed to solve the self-consistent equations. For the problem at hand, however, analytic solutions can be constructed in the vicinity of  $\omega=0$   $(E_F)$ . To facilitate this treatment, it is useful to consider the large  $\Sigma$  limit of the Green function:

$$G(\omega) = \int \frac{g(\epsilon)d\epsilon}{\omega - \Sigma - \epsilon}$$

$$= -\frac{1}{\Sigma} \left( 1 + \frac{\omega - a_1}{\Sigma} + \frac{a_2 - 2a_1\omega + \omega^2}{\Sigma^2} \right), \quad (10)$$

where  $a_1 = \int \epsilon g(\epsilon) d\epsilon = 0$  in our case and  $a_2 = \int \epsilon^2 g(\epsilon) d\epsilon$ . It then follows that Eq. (6) can be approximated as

$$\xi(\omega) \approx \omega - a_2 G(\omega).$$
 (11)

It should also be noticed that for a symmetric classical density of states  $P(\epsilon) = P(-\epsilon)$ , all quantities  $\Sigma$ , G, and  $\xi$  are purely imaginary at  $\omega = 0$ .

#### III. ANALYTICAL SOLUTIONS AROUND $\omega = 0$

#### A. d = 3 systems

As remarked earlier,  $\Sigma$ , G, and  $\xi$  are purely imaginary when  $\omega = 0$ . Consider now the  $\omega = 0$  limit of Eq. (8) for d = 3. In this limit, Eq. (8) becomes

$$G_2(\omega=0) = -\frac{3}{B} \left(\frac{\xi_2}{B}\right)^2 \left(\frac{B}{\xi_2} - \arctan\frac{B}{\xi_2}\right). \tag{12}$$

Noting from Eq. (11) that  $\xi_2 = -a_2G_2$ , we obtain

$$\xi_2 \left[ 1 - \frac{B^2}{3a_2} - \frac{\xi_2}{B} \arctan \frac{B}{\xi_2} \right] = 0 .$$
 (13)

One immediately sees that for  $\frac{3a_2}{B^2} < 1$ , the only solution is  $\xi_2(\omega = 0) = 0$ . This implies that  $G_2 = 0$  and therefore  $\rho(0) = 0$ . For  $\frac{3a_2}{B^2} > 1$ , a finite solution exists for  $\xi_2$ ,

$$\xi_2 = -\frac{B(3a_2 - B^2)}{3a_2 \arctan\frac{3a_2\pi}{2(3a_2 - B^2)}} \tag{14}$$

and the density of states is simply given by  $\rho(0) = -\frac{\xi_2}{a_2\pi}$ . An expansion around  $\omega = 0$  for  $\frac{3a_2}{B^2} < 1$  yields

$$\xi = \frac{\omega}{1 - 2Bca_2} + ic\pi a_2 \left(\frac{\omega}{1 - 2Bca_2}\right)^2. \tag{15}$$

Hence,

$$\rho(\omega) = P\left(\frac{\omega}{1 - 2Bca_2}\right). \tag{16}$$

We see then that quantum hopping renormalizes the gap parameter when the hopping integral is small. This conclusion was obtained in Ref. 5 as well. Equation (16) is plotted in Fig. 1. As is evident a significant increase in the density of states occurs only if t/B > 0.167. In calculating the values of  $a_2$ , we have used a flat density of states for  $g(\epsilon)$  with half-width 2dt.

# B. d=2 systems

Consider now the case of d=2. From Eq. (9), we observe that

$$G_2(\omega = 0) = -\frac{\xi_2}{B^2} \ln \frac{B^2 + \xi_2^2}{\xi_2^2}$$
 (17)

Again, using the relation  $\xi_2 = -a_2G_2$ , we obtain

$$\xi_2(\omega = 0) = -\frac{B}{(e^{\frac{B^2}{a_2}} - 1)^{1/2}} \approx -Be^{-\frac{B^2}{2a_2}}$$
 (18)

and therefore

$$\rho(0) = \frac{B}{a_2 \pi} e^{-\frac{B^2}{2a_2}}. (19)$$

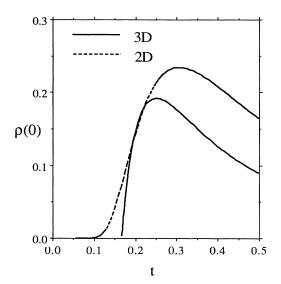


FIG. 1. The density of states (in units of  $\frac{1}{Ba^3}$ ) at  $\omega=0$  as a function of the hopping integral (in units of B). B is the Coulomb gap width and a is the lattice constant. For d=3,  $\rho(0)=0$  for  $t/B<\frac{1}{6}$ . A flatband is used for the unperturbed density of states.

Although the density of states is nonzero, it is exponentially small for small values of t. If  $\xi$  is small in the vicinity of  $\omega = 0$ , we find that

$$G(\xi) = \frac{\xi}{B} \left[ -\ln B^2 \xi_1^2 + \xi_2^2 - 2i \arctan \frac{\xi_1}{\xi_2} \right]$$
 (20)

or equivalently,

$$\xi = \frac{B^2 \omega}{B^2 - a_2 \ln \frac{B^2}{\xi_1^2 + \xi_2^2} - i2a_2 \arctan \frac{\xi_1}{\xi_2}} \ . \tag{21}$$

Two limiting cases of Eq. (21) are of interest here.

(a) If  $\omega < \frac{2a_2^2\pi\rho(0)}{R^2}$ , then

$$\rho(\omega) = \rho(0) \left[ 1 + \left( \frac{B\omega}{2a_2} \right)^2 e^{-\frac{B^2}{2a_2}} \right]. \tag{22}$$

This form is valid only for exponentially small  $\omega$  for small t.

(b) The other case corresponds to  $\frac{2a_2^2}{B^2} << 1$  and  $\omega > Be^{-\frac{B^2}{2a_2}}$ , in which case

$$\rho(\omega) = P\left(\frac{\omega}{1 - \frac{4a_2}{B^2} \ln \frac{B}{|\omega|}}\right). \tag{23}$$

Again, the effect of quantum hopping is seen as a renormalization of the gap parameter. However, the renormalization factor increases with decreasing  $\omega$ .

The density of states at  $\omega=0$  for d=2 is indicated with the dashed line in Fig. 1. It is evident that only when t reaches a value  $t_1$ ,  $t_1/B=0.12$ , does the density of states at  $\omega=0$  become significant.

Equation (19) was interpreted by Vignale and coworkers as evidence for the absence of a Coulomb gap in two dimensions. We see that on a closer look, that conclusion does not hold. An exponentially small density of states in a small region around the Fermi energy should not affect any conclusions made on the Coulomb gap account for transport properties. Equation (23) definitely shows that when the hopping integral is small, the Coulomb gap persists even in two dimensions.

# IV. NUMERICAL RESULTS

To obtain a solution for general  $\omega$ , we employed the following numerical iteration scheme.

- (a) Supply initial values for  $\Sigma$  and G and solve for  $\xi = \Sigma + 1/G$ .
- (b) Obtain a new value of G, G', from the CPA equation [Eqs. (8) and (9) for d = 3 and d = 2, respectively];

$$G'=G_d(\mathcal{E})$$

(c) Update the value of  $\xi$  from the inverse of the unperturbed Green function,

$$\xi' = \omega + \frac{1}{G'} - Z_0(G')$$
,

where  $Z_0(G)$  is the inverse function of  $G_0(Z)$ , i.e.,  $G(Z_0(G)) = G$ .

(d) Repeat steps (b) and (c) until the solution converges. In our calculation, we used a flat density of states

of half-width 2dt for the unperturbed Green function, <sup>11</sup>

$$G_0(Z) = \frac{1}{4dt} \ln \frac{Z + 2dt}{Z - 2dt}$$
 (24)

and the inverse function

$$Z_0(G) = 2dt \operatorname{cotanh}(2dtG). \tag{25}$$

The density of states for various values of t is plotted in Fig. 2 for d=3 and in Fig. 3 for d=2. As in the  $\omega=0$  case, we find that a significant increase in the low energy density of states occurs only for t/B>0.1 for d=2 and t/B>0.167 for d=3.

# V. DISCUSSIONS AND CONCLUSIONS

To determine how significant the increase in the density of states is in response to quantum tunneling, we must estimate the magnitude of the hopping integral corresponding to t/B>0.1. The ratio of  $\frac{t}{B}$  pertinent to experiment can be estimated for d=3 as follows. B is given by  $\frac{ce^2}{\kappa r}$ , where  $r=n^{1/3}$  is the interimpurity distance,  $\kappa$  the dielectric constant, and c is a constant of order unity. According to Mott, c the hopping integral is given by

$$t = \frac{e^2}{\kappa a_H} \left( 1 + \frac{r}{a_H} \right) e^{-\frac{r}{a_H}} , \qquad (26)$$

where  $a_H$  is the orbital radius of an isolated impurity. The ratio t/B,

$$\frac{t}{B} = \frac{y(1+y)}{c}e^{-y} , (27)$$

can be expressed in terms of the scaled concentration  $y=4.0(\frac{n}{n_c})^{1/3}$ , where  $n_c$  is the critical concentration at the metal-insulator transition. In obtaining Eq. (27), we used the Mott criterion<sup>12</sup>

$$n_c^{1/3} a_H = 0.25 (28)$$

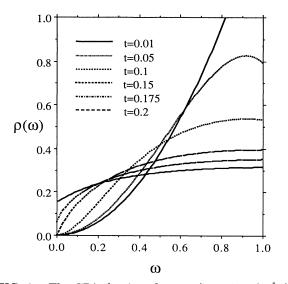


FIG. 2. The CPA density of states (in units of  $\frac{1}{Ba^3}$ ) of a three dimensional system for different values of hopping integral (in units of B).

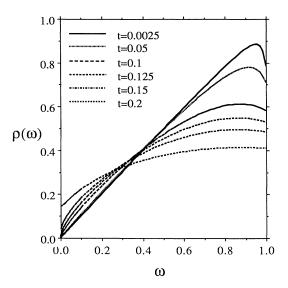


FIG. 3. The CPA density of states (in units of  $\frac{1}{Ba^3}$ ) of a two dimensional system for different values of hopping integral (in units of B).

which is known to hold for a wide range of materials. The ratio t/B is graphed in Fig. 4 where t/B has been normalized to 1 at  $n_c$  to eliminate c. In almost all of the experiments in which hopping conduction is observed, the impurity concentration is less than  $n_c/3$ . We see from Fig. 4 that at  $n = n_c/3$ , t/B drops to approximately 30%. If all states are Anderson localized in d = 3, then t/B cannot exceed a critical value. This critical value depends on the details of the system considered. For a flat distribution with half-width W, the critical disorder strength is known to be  $W_c/t = 7.8.^{13}$  An estimate for the critical value in our problem can be obtained by requiring

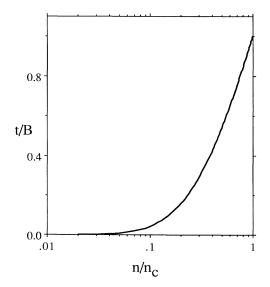


FIG. 4. The estimated ratio of the hopping integral to the gap width as a function of the doping concentration. t/B has been normalized to 1 at the critical concentration of the M-I transition,  $n_G$ .

that the second moment of the disorder equals that for a flat distribution. This yields a critical ratio  $\frac{t_c}{B}=0.16$  at  $n_c$ . It follows then that the relevant experimental range corresponds to t/B<0.05.

An alternative way of estimating the ratio appropriate to experiment is to calculate the localization length. The localization length varies as  $L_c = L_c(0)(1-\frac{n}{n_c})^{-\nu}$  when close to the metal-insulator transition.  $L_c(0) \approx a_H$ . The exponent  $\nu$  ranges from 0.5 to 1.<sup>13</sup> It then follows that

$$L_c/r = 0.25 \left(\frac{n_c}{n}\right)^{1/3} \left(1 - \frac{n}{n_c}\right)^{-\nu}.$$
 (29)

For  $n/n_c = 0.3$ , Eq. (29) yields a value of  $L_c/r = 0.19$  if  $\nu = 0.5$ . Alternatively,  $L_c/r = 0.22$  if  $\nu = 1$ . We require, therefore, that the values of t/B be chosen such that a calculation of the localization length with the Hamiltonian Eq. (1) yields the correct value of the localization length at the Fermi level; that is,  $L_c/r = 0.2$ . We have carried out such a calculation using the Green function method.<sup>13</sup> From this calculation, we find again that the ratio t/B = 0.05 corresponds to  $L_c/r = 0.2$ . The argument outlined above cannot be directly extended to d=2systems because no Anderson transition is expected in d=2. However, the values of t and B for a given concentration are not expected to depend on the dimensionality because t describes the wave-function overlap and B the strength of the long-range interaction. These quantities should be determined primarily by the type of material, the dopant, and the impurity concentration. We propose, therefore, that the bound t/B < 0.05 should also apply to d=2 systems.

In our calculations, however, values of t/B > 0.1 were needed to fill in the Coulomb gap at T=0. Consequently, quantum hopping does not significantly alter the soft Coulomb gap. For t/B=0.05, the increase in the density of states in d=3 is only about 5%. The same

is true for d=2 and  $\omega>0.03B$ . The larger relative increase in the density of states at very low energy is irrelevant to hopping conduction in the experimental temperature range. This is because in the theory of Efros and Shklovskii, the hopping conductance of an interacting system is evaluated from the single particle density of states.<sup>2</sup> The hopping conductivity is determined by a critical conductance  $G_c$  such that the links with conductance larger than  $G_c$  form a percolating network that connects the two leads. A simple estimate shows that the contribution from  $\omega<0.01B$  to the conduction process is small in experiments as long as conduction is controlled by the Coulomb gap. The Coulomb gap, therefore, remains intact for both two and three dimensional systems in the presence of quantum tunneling.

In conclusion, the effect of the quantum hopping on the single particle density of states in the Coulomb gap problem has been studied. Although there is an increase of density of states at low energy, the effect is too small to affect hopping conduction within the single electron hopping picture in the parameter range pertinent to experiment, for both d=2 and d=3 dimensional systems. Significant increase in the density of states and the filling-in of the gap through quantum hopping can only occur when the system is in the critical region of the metal-insulator transition, where the localization length becomes large and quantum effects begin to dominate. It remains an open question then why the noninteracting form of variable range hopping conductivity was observed by Timp  $et\ al.^7$ 

# ACKNOWLEDGMENTS

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