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Self-Consistent Pair Correlations in Mott-Type Insulators

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A simple cubic hydrogenic lattice with the Hubbard Hamiltonian is used as a model for Mott-type insulators. A new self-consistent approximation is developed by the decoupling of four-particle Green's functions, leading to a higher-order Hartree-like eigenvalue equation for electron-hole Green's functions. This equation is an improvement over the previously used Ω approximation. The solution of the eigenvalue equation leads to the conjecture that an exact solution (i.e., one without decoupling) would result in each atom having exactly one electron.

I. INTRODUCTION

THE properties of most insulators can be explained by conventional energy-band theory. There are some exceptions to this rule. In this paper another model will be investigated.

Mott[1] has called attention to the fact that metal atoms at large separations should constitute an insulator. A model of these Mott-type insulators has been investigated by Hubbard[2] and by Kemeny.[3–5] Both authors used Zubarev[6] two-time Green's functions. Hubbard employed the one-electron, and Kemeny the two-particle electron-hole variety. The latter approach shows that under appropriate circumstances bound electron-hole pairs appear, which is the original picture of Mott.

In I we defined the retarded electron-hole Green's function of the Zubarev type.

$$G_{ijj'i'}(t-t')=\langle\langle c_{i\sigma}{}^{\dagger}(t)c_{j\sigma}(t)\,;\,c_{j'\sigma}{}^{\dagger}(t')c_{i'\sigma}(t')\rangle\rangle\,. \qquad (1.1)$$

The equation of motion for this Green's function contained a higher-order correlation term

$$J\langle\langle(n_{j-\sigma}-n_{i-\sigma})c_{i\sigma}{}^{\dagger}c_{j\sigma}\,;\,c_{j'\sigma}{}^{\dagger}c_{i'\sigma}\rangle\rangle\,. \qquad (1.2)$$

We incorporated $\langle(n_{j-\sigma}-n_{i-\sigma})\rangle$ into an effective potential, still denoted by J, without giving a prescription as to how it should be calculated. Thus the calculation was not self-consistent.

The primary purpose of this paper is the derivation and solution of a self-consistent equation valid in the insulating range. This will be done using the Martin-Schwinger[7] many-time Green's-function technique. In the insulating range, all electron-hole pairs are bound. Such a situation can be treated by considering two-particle electron-hole Green's functions. It is not even necessary to solve for the entire electron-hole Green's function; it is enough to obtain its homogeneous part. This is obvious from I, where we found that bound states were characterized by the homogeneous part of the equation for the spectral representation.

II. TRANSFORMATION OF THE HAMILTONIAN

In I we used the Hubbard Hamiltonian[2]

$$H=\sum_{ij}\sum_{\sigma}T_{ij}c_{i\sigma}{}^{\dagger}c_{j\sigma}+J\sum_{i}n_{i\sigma}n_{i-\sigma}\,. \qquad (2.1)$$

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[1] N. F. Mott, Proc. Phys. Soc. (London) **62**, 416 (1949); Advan. Phys. **13**, 325 (1964).

[2] J. Hubbard, Proc. Roy. Soc. (London) **A276**, 238 (1961); **A281**, 401 (1964).

[3] G. Kemeny, Ann. Phys. (N. Y.) **32**, 69 (1965), hereafter referred to as I.

[4] G. Kemeny, Ann. Phys. (N. Y.) **32**, 404 (1965).

[5] G. Kemeny, Phys. Letters **14**, 87 (1965).

[6] D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

[7] P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1342 (1959).

The first term describes the hopping motion of the electrons from atom to atom. The second one is the repulsion of two electrons on the same atom.

However, when dealing with Martin-Schwinger Green's functions it is customary to work with a slightly modified Hamiltonian

$$H'=\sum_{ij}\sum_{\sigma} T_{ij}c_{i\sigma}{}^{\dagger}c_{j\sigma}+J\sum_{i} n_{i\sigma}n_{i-\sigma}-\mu\sum_{i}\sum_{\sigma} n_{i\sigma}, \quad (2.2)$$

where we have added the chemical potential term to the original form (2.1). This is done in order to ensure the proper boundary condition on the Green's functions.

In order to go into the electron-hole representation which is best suited for this problem, we introduce a transformation[5] into this Hamiltonian. The positive-spin electron operators are left intact, but the negative-spin electron operators are transformed into hole operators:

$$\begin{aligned} c_{i+}{}^{\dagger}&=\psi_{i+}{}^{\dagger}, & c_{i-}{}^{\dagger}&=\psi_{i-}, \\ c_{i+}&=\psi_{i+}, & c_{i-}&=\psi_{i-}{}^{\dagger}. \end{aligned} \quad (2.3)$$

This yields

$$\begin{aligned} H'=\sum_{ij} T_{ij}\psi_{i+}{}^{\dagger}\psi_{j+}-\sum_{ij} T_{ij}\psi_{i-}{}^{\dagger}\psi_{j-}-J\sum_{i} n_{i+}n_{i-} \\ +J\sum_{i} n_{i+}-\mu\sum_{i} n_{i+}+\mu\sum_{i} n_{i-}, \quad (2.4) \end{aligned}$$

apart from unnecessary additive constants, where n_{i-} now is the number of holes in the negative-spin states. Since we wish to calculate for the Mott transition, we are only interested in the case where there is an average of one electron per site with as many up-spin electrons as there are down-spin holes. This implies that the Hamiltonian must be symmetric with respect to n_{i+} and n_{i-}. Consequently

$$\mu=J/2,$$

and the Hamiltonian becomes, to within a constant,

$$\begin{aligned} H'=\sum_{ij} T_{ij}\psi_{i+}{}^{\dagger}\psi_{j+}-\sum_{ij} T_{ij}\psi_{i-}{}^{\dagger}\psi_{j-} \\ -J\sum_{i} n_{i+}n_{i-}+\tfrac{1}{2}J\sum_{i} n_{i+}+\tfrac{1}{2}J\sum_{i} n_{i-}. \quad (2.5) \end{aligned}$$

The repulsive electron-electron interaction has been transformed into the attractive electron-hole interaction at the cost of changing the one-particle energy.

III. GREEN'S FUNCTIONS

We introduce the Martin-Schwinger many-time Green's functions[7]

$$\begin{aligned} &G_n(12\cdots n;1'2'\cdots n') \\ &\equiv(-i)^n\epsilon\langle(\psi(1)\psi(2)\cdots\psi(n)\psi^{\dagger}(n')\cdots \\ &\qquad\times\psi^{\dagger}(2')\psi^{\dagger}(1'))\rangle_{+}, \quad (3.1) \end{aligned}$$

where $1, 2\cdots n$ stand for the space-time and temporarily for the spin arguments of the field operators. The positive subscript indicates the time ordering and ϵ is the antisymmetrical function of the time arguments.

As we have mentioned in the Introduction, we need an equation only for the homogeneous part of the electron-hole Green's function. An equation containing also the inhomogeneous terms was given by Martin and Schwinger. Omitting these inhomogeneous parts, this equation appears in the form

$$\begin{aligned} \left[i\frac{\partial}{\partial t_1}-h_1\right]\left[i\frac{\partial}{\partial t_2}-h_2\right]G_2(12;1'2')-iv(12)G_2(12;1'2') \\ =-\int\!\!\int d3d4v(13)v(24)G_4(1234;1'2'3^+4^+). \quad (3.2) \end{aligned}$$

For the present problem

$$v(12)=-J\delta(1-2), \quad (3.3)$$

$$\begin{aligned} h_{1\pm}G_2(12;1'2')=\tfrac{1}{2}JG_2(12;1'2') \\ \pm\sum_{I} T_{1I}G_2(I2;1'2'), \quad (3.4) \end{aligned}$$

where 1 and I have equal-time components. In the Ω approximation, introduced by Martin and Schwinger, the right-hand side of Eq. (3.2) is neglected. This corresponds to the procedure adopted in I. The self-consistency requirement means that G_4 is to be retained.

Specializing Eq. (3.2) to the model at hand leads to

$$\begin{aligned} (G_{1_+}{}^0)^{-1}(G_{2_-}{}^0)^{-1}G_2(1_+2_-;1_+{}'2_-{}') \\ +iJ\delta(1-2)G_2(1_+2_-;1_+{}'2_-{}') \\ =-J^2G_4(1_+2_-1_-2_+;1_+{}'2_-{}'1_-{}^+2_+{}^+), \quad (3.5) \end{aligned}$$

where

$$(G^0)^{-1}=i\frac{\partial}{\partial t}-h. \quad (3.6)$$

If $G_2(1_+2_-;1_+{}'2_-{}')$ indeed expresses a bound state, a stable finite solution is expected even if the temporal separation of the primed from the unprimed indices is very large, provided that the primed indices, and also the unprimed ones, are near each other in time.[8] Utilizing this idea for decoupling G_4, we infer

$$\begin{aligned} &\{(G_{1_+}{}^0)^{-1}(G_{2_-}{}^0)^{-1}+iJ\delta(1-2)\}G_2(1_+2_-;1_+{}'2_-{}') \\ &\quad=-J^2\{G_2(1_+2_-;1_+{}'2_-{}')G_2(1_-2_+;1_-{}^+2_+{}^+) \\ &\qquad+G_2(1_+2_-;2_+{}^+1_-{}^+)G_2(1_-2_+;2_-{}'1_+{}') \\ &\qquad-G_2(1_+1_-;1_+{}'2_-{}')G_2(2_-2_+;1_-{}^+2_+{}^+) \\ &\qquad-G_2(1_+1_-;2_+{}^+1_-{}^+)G_2(2_-2_+;2_-{}'1_+{}')\}. \quad (3.7) \end{aligned}$$

The right-hand side of this equation describes a Hartree-Fock–type approximation for two particles. An electron and a hole proceed from $1_+{}'2_-{}'$ to 1_+2_- directly in the first term, with one particle exchanged in the last two terms each, and both particles exchanged in the second term. A straight Hartree approximation would neglect all but the direct term. This would, however, violate the condition inherent in Eq. (3.5) that the right-hand side should vanish for $1=2$. It should be recognized that the last two terms should be partially retained in the Hartree approximation since only one of the particles is

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[8] L. P. Kadanoff and P. C. Martin, Phys. Rev. **124**, 670 (1961).

exchanged. Because of symmetry of 1 and 2, one may retain either of the last two terms. The Hartree approximation thus gives

$$\{(G_{1+}{}^0)^{-1}(G_{2-}{}^0)^{-1}+iJ\delta(1-2)\}G_2(1_+2_-;1_+{}'2_-{}')$$
$$=-J^2\{G_2(1_+2_-;1_+{}'2_-{}')G_2(1_-2_+;1_-{}^+2_+{}^+)$$
$$-G_2(1_+1_-;1_+{}'2_-{}')G_2(2_-2_+;1_-{}^+2_+{}^+)\}. \quad (3.8)$$

Let us examine the homogeneous part of

$$G_2(1_+2_-;1_+{}^+2_-{}^+)=-\langle[n(1_+)-\langle n(1_+)\rangle]$$
$$\times[n(2_-)-\langle n(2_-)\rangle]\rangle, \quad (3.9)$$

or more specifically of

$$G_2(1_+1_-;1_+{}^+1_-{}^+)=-\langle[n(1_+)-\langle n(1_+)\rangle]$$
$$\times[n(1_-)-\langle n(1_-)\rangle]\rangle. \quad (3.10)$$

Since the interaction is attractive, the correlation is positive, i.e., $G_2(1_+1_-;1_+{}'1_-{}')$ is real and negative. Using the Gor'kov factorization[8,9] formulas

$$F(1_+2_-)=-i\epsilon\langle\psi(1_+)\psi(2_-)\rangle, \quad (3.11)$$

$$F^+(1_+2_-)=-i\epsilon\langle\psi^+(1_+)\psi^+(2_-)\rangle, \quad (3.12)$$

we find

$$G_2(1_+1_-;1_+{}^+1_-{}^+)=-F(1_+1_-)F^+(1_+1_-). \quad (3.13)$$

Since the left-hand side is real and negative,

$$F^+(1_+1_-)=F^*(1_+1_-). \quad (3.14)$$

Therefore the Gor'kov factorized G_2 should have the form

$$G_2(1_+2_-;1_+{}'2_-{}')=-F(1_+2_-)F^*(1_+{}'2_-{}'). \quad (3.15)$$

Using this factorization procedure in Eq. (3.8), we get

$$\{(G_{1+}{}^0)^{-1}(G_{2-}{}^0)^{-1}+iJ\delta(1-2)\}F(1_+2_-)$$
$$=J^2\{F(1_+2_-)F(1_-2_+)$$
$$-F(1_+1_-)F(2_-2_+)\}F^*(1_-2_+). \quad (3.16)$$

We seek a solution of these equations which corresponds to the ground state of a bound pair. We wish to convert these equations into eigenvalue equations with a time-independent effective potential for $t_1=t_2$. Thus we require

$$F(1_+2_-)=F^*(1_-2_+), \quad (3.17)$$

which in view of the lattice- and time-translational invariances leads to

$$[(G_{1+}{}^0)^{-1}(G_{2-}{}^0)^{-1}+iJ\delta(1-2)]F(1_+2_-)$$
$$=J^2\{|F(1_+2_-)|^2-|F(1_+1_-)|^2\}F(1_+2_-). \quad (3.18)$$

IV. EIGENVALUE EQUATION

Following Martin and Schwinger[7] we utilize the operator identity

$$G_{1+}{}^0G_{2-}{}^0=[(G_{1+}{}^0)^{-1}+(G_{2-}{}^0)^{-1}]^{-1}(G_{1+}{}^0+G_{2-}{}^0). \quad (4.1)$$

The unperturbed electron Green's function is

$$G_{1+}{}^0(\mathbf{R}_1t_1;\mathbf{R}_1{}'t_1{}')=\tfrac{1}{2}\{\tan(h_{1+}\tau/2)-i\epsilon(t_1-t_1{}')\}$$
$$\times e^{-ih_{1+}(t_1-t_1{}')}\delta_{\mathbf{R}_1\mathbf{R}_1{}'}, \quad (4.2)$$

where

$$\tau=-i\beta, \quad (4.3)$$

and

$$\epsilon(t_1-t_1{}')=1 \quad \text{for} \quad t_1>t_1{}'$$
$$=-1 \quad \text{for} \quad t_1<t_1{}', \quad (4.4)$$

and a similar expression stands for $G_{2-}{}^0$. We also note that

$$(G_{1+}{}^0)^{-1}+(G_{2-}{}^0)^{-1}=i\left(\frac{\partial}{\partial t_1}+\frac{\partial}{\partial t_2}\right)-h_{1+}-h_{2-}. \quad (4.5)$$

Equation (4.1) can be transformed into

$$[(G_{1+}{}^0)^{-1}+(G_{2-}{}^0)^{-1}]G_{1+}{}^0G_{2-}{}^0=G_{1+}{}^0+G_{2-}{}^0. \quad (4.6)$$

This expression can be utilized for the integration of Eq. (3.18) and leads to

$$[(G_{1+}{}^0)^{-1}+(G_{2-}{}^0)^{-1}]F(\mathbf{R}_{1+}t_1\mathbf{R}_{2-}t_2)=\int_0^\tau\!\!\int d\bar{1}d\bar{2}[G_{1+}{}^0(1-\bar{1})+G_{2-}{}^0(2-\bar{2})]$$
$$\times\{-iJ\delta(\bar{1}-\bar{2})+J^2[|F(\bar{\mathbf{R}}_{1+}{}'\bar{\mathbf{R}}_{2-}{}')|^2-|F(\bar{\mathbf{R}}_{1+}{}'\bar{\mathbf{R}}_{1-}{}')|^2]\}F(\bar{1}_+\bar{2}_-). \quad (4.7)$$

Substituting the Fourier-series expression with the required periodicity property

$$F(\mathbf{R}_{1+}t_1\mathbf{R}_{2-}t_2)=\frac{1}{\tau}\sum_{\nu'}e^{i\pi\nu'(t_1+t_2)/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu'), \quad \nu'=\text{odd}, \quad (4.8)$$

into Eq. (4.7) and using the expression for the unperturbed electron Green's function in Eq. (4.2), we get

$$\sum_{\nu'}\left[i\left(\frac{\partial}{\partial t_1}+\frac{\partial}{\partial t_2}\right)-h_{1+}-h_{2-}\right]e^{i\pi\nu'(t_1+t_2)/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu')$$
$$=\sum_{\nu'}\int_0^\tau\!\!\int d\bar{1}d\bar{2}\{\tfrac{1}{2}[\tan(h_{1+}\tau/2)-i\epsilon(t_1-t_1{}')]e^{-ih_{1+}(t_1-t_1{}')}\delta_{\mathbf{R}_1\bar{\mathbf{R}}_1}\delta(2-\bar{2})+\tfrac{1}{2}[\tan(h_{2-}\tau/2)-i\epsilon(t_2-t_2{}')]$$
$$\times e^{-ih_{2-}(t_2-t_2{}')}\delta_{\mathbf{R}_2\bar{\mathbf{R}}_2}\delta(1-\bar{1})\}\{-iJ\delta(\bar{1}-\bar{2})+J^2[|F(\bar{\mathbf{R}}_1\bar{\mathbf{R}}_2)|^2-F(\bar{\mathbf{R}}_1\bar{\mathbf{R}}_1)|]^2\}e^{i\pi\nu'(t_1{}'+t_2{}')/\tau}F(\bar{\mathbf{R}}_{1+}\bar{\mathbf{R}}_{2-}\nu'). \quad (4.9)$$

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[9] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **34**, 735 (1958) [English transl.: Soviet Phys.—JETP **7**, 505 (1958)].

The left-hand side term is then equal to

$$\sum_{\nu'}(-2\pi\nu'/\tau-h_{1+}-h_{2-})e^{i\pi\nu'(t_1+t_2)/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu'). \tag{4.10}$$

Integration of the J term on the right-hand side yields

$$-iJ\sum_{\nu'}\{\tfrac{1}{2}[\tan(h_{1+}\tau/2)-i\epsilon(t_1-t_2)]e^{-ih_{1+}(t_1-t_2)}+\tfrac{1}{2}[\tan(h_{2-}\tau/2)-i\epsilon(t_2-t_1)]e^{-ih_{2-}(t_2-t_1)}\}$$
$$\times e^{i\pi\nu'(t_1+t_2)/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu')\delta\mathbf{R}_{1+}\mathbf{R}_{2-}, \tag{4.11}$$

whereas integration of the J^2 term on the right-hand side gives

$$J^2[|F(\mathbf{R}_1\mathbf{R}_2)|^2-|F(\mathbf{R}_1\mathbf{R}_1)|^2]\sum_{\nu'}\left\{\frac{1}{2}\left[(\tan(h_{1+}\tau/2))\frac{(e^{-i(\pi\nu'+h_{1+}\tau)}-1)}{i(\pi\nu'/\tau+h_{1+})}+\frac{(e^{i(\pi\nu'+h_{1+}\tau)}-2e^{i(\pi\nu'/\tau+h_{1+})t}+1)}{(\pi\nu'/\tau+h_{1+})}\right]\right.$$
$$\times e^{-i(h_{1+}-\pi\nu'/\tau)t}+\frac{1}{2}\left[(\tan(h_{2-}\tau/2))\frac{(e^{-i(\pi\nu'+h_{2-}\tau)}-1)}{i(\pi\nu'/\tau+h_{2-})}\right.$$
$$\left.\left.+\frac{(e^{i(\pi\nu'+h_{2-}\tau)}-2e^{i(\pi\nu'/\tau+h_{2-})t}+1)}{(\pi\nu'/\tau+h_{1+})}\right]e^{-i(h_{2-}-\pi\nu'/\tau)t}\right\}e^{i\pi\nu'(t_1+t_2)/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu'). \tag{4.12}$$

The

$$ih\tau=h\beta \tag{4.13}$$

argument arises here from the G^0. These describe the motion of particles in the unperturbed system, i.e., electrons above and holes below the Fermi level. For both particles $h>0$. Moreover, since we seek the solution only for $t_1=t_2=t$, Eq. (4.9) becomes at $T=0$

$$\sum_{\nu'}(-2\pi\nu'/\tau-h_{1+}-h_{2-})e^{i2\pi\nu't/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu')$$
$$=-J\sum_{\nu'}e^{i2\pi\nu't/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu')\delta\mathbf{R}_{1+}\mathbf{R}_{2-}-J^2[|F(\mathbf{R}_1\mathbf{R}_2)|^2-|F(\mathbf{R}_1\mathbf{R}_1)|^2]$$
$$\times\sum_{\nu'}\left[\frac{1}{(\pi\nu'/\tau+h_{1+})}+\frac{1}{(\pi\nu'/\tau+h_{2-})}\right]e^{i2\pi\nu't/\tau}F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu'). \tag{4.14}$$

Multiplying by $\tau^{-1}e^{-i2\pi\nu t/\tau}$ and integrating over t, we get

$$(-2\pi\nu/\tau-h_{1+}-h_{2-})F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu)=-JF(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu)\delta\mathbf{R}_{1+}\mathbf{R}_{2-}-J^2[|F(\mathbf{R}_1\mathbf{R}_2)|^2-|F(\mathbf{R}_1\mathbf{R}_1)|^2]$$
$$\times\left[\frac{1}{(\pi\nu/\tau+h_{1+})}+\frac{1}{(\pi\nu/\tau+h_{2-})}\right]F(\mathbf{R}_{1+}\mathbf{R}_{2-}\nu). \tag{4.15}$$

Replacing $2\pi\nu/\tau$ by the continuous variable ω we obtain the eigenvalue equation

$$(\omega+h_{1+}+h_{2-})F(\mathbf{R}_{1+}\mathbf{R}_{2-}\omega)=JF(\mathbf{R}_{1+}\mathbf{R}_{2-}\omega)\delta\mathbf{R}_{1+}\mathbf{R}_{2-}+J^2[|F(\mathbf{R}_1\mathbf{R}_2)|^2-|F(\mathbf{R}_1\mathbf{R}_1)|^2]$$
$$\times\left[\frac{1}{(\omega/2+h_{1+})}+\frac{1}{(\omega/2+h_{2-})}\right]F(\mathbf{R}_{1+}\mathbf{R}_{2-}\omega). \tag{4.16}$$

Since this is a nonlinear equation, the normalization of its eigenfunction is very important. The normalization can be determined from the case in which J is the dominating influence in the energy. In that case only $F(1_+1_-)$ is different from zero in the ground state because of the extremely strong binding of the pair. From Eqs. (3.10) and (3.15) we find that

$$|F(1_+1_-)|^2=\langle n(1_+)n(1_-)\rangle-\langle n(1_+)\rangle\langle n(1_-)\rangle. \tag{4.17}$$

The probability of having any particular state occupied is $\frac{1}{2}$. Thus $\langle n(1_+)\rangle=\langle n(1_-)\rangle=\frac{1}{2}$. Because of the extremely strong binding, there is either an electron in the positive-spin state and a hole in the negative one, or vice versa. Thus $\langle n(1_+)n(1_-)\rangle=\frac{1}{2}$ and

$$|F(1_+1_-)|^2=\tfrac{1}{4}. \tag{4.18}$$

We conclude that

$$\sum_{\mathbf{R}_1}|F(\mathbf{R}_{1+}\mathbf{R}_{2-})|^2=\sum_{\mathbf{R}_2}|F(\mathbf{R}_{1+}\mathbf{R}_{2-})|^2=\tfrac{1}{4} \tag{4.19}$$

is the correct normalization. If $F(\mathbf{R}_{1+}\mathbf{R}_{2-})$ is to be considered the wave function of the pair, it is preferable to normalize it to unity. Adopting this normalization, we write the equation as

$$[\omega+h_{1+}+h_{2-}-J\delta_{\mathbf{R}_1\mathbf{R}_2}]\psi(R_{1+}R_{2-})$$

$$=\tfrac{1}{4}J^2[|\psi(\mathbf{R}_1\mathbf{R}_2)|^2-|\psi(\mathbf{R}_1\mathbf{R}_1)|^2]$$

$$\times\left[\frac{1}{(\omega/2+h_{1+})}+\frac{1}{(\omega/2+h_{2-})}\right]\psi(\mathbf{R}_{1+}\mathbf{R}_{2-}), \quad (4.20)$$

with the conventional notation for wave functions. This is a self-consistent field equation to determine the pair wave function and eigenvalue. In contradistinction to the conventional situation, it is quadratic in the operators and in the eigenvalue. This is due to the fact that the potential field felt by a pair depends on the motion of another pair governed by the same dynamics.

According to the assumption in Eq. (4.19),

$$|\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})|^2=|\psi(\mathbf{R}_{1-}\mathbf{R}_{2+})|^2. \quad (4.21)$$

This is the reason for omitting spin indices in the self-consistent potential of Eq. (4.20). If the spin indices are chosen here as on the right of Eq. (4.21) a physical interpretation emerges. Equation (4.20) is a Schrödinger equation for the $(\mathbf{R}_{1+}\mathbf{R}_{2-})$ pair. The effective attractive interaction between the members of this pair depends on the correlated opposite spin populations on these atoms, i.e., on the $(\mathbf{R}_{1-}\mathbf{R}_{2+})$ pair. If the latter pair is concentrated on one atom, i.e.,

$$|\psi(\mathbf{R}_{1-}\mathbf{R}_{1+})|^2=1, \quad |\psi(\mathbf{R}_{1-}\mathbf{R}_{2+})|^2=0, \quad (4.22)$$

the interaction is the strongest. If this pair is weakly bound, i.e.,

$$|\psi(\mathbf{R}_{1-}\mathbf{R}_{1+})|^2\approx|\psi(\mathbf{R}_{1-}\mathbf{R}_{2+})|^2, \quad (4.23)$$

the interaction is weak. The two pairs are in a mutually symmetrical situation and both are either strongly or weakly bound in their ground states. If a pair is concentrated on one atom, the intrusion of either member of another pair requires the expenditure of potential energy to drive one member of the first pair off this atom. This can be accomplished only if sufficient band energy is gained in the process.

V. SOLUTION OF THE EIGENVALUE EQUATION

The eigenvalue equation (4.20) can be brought to the form

$$\left[1-\frac{J\delta_{\mathbf{R}_1\mathbf{R}_2}}{(\omega+h_{1+}+h_{2-})}\right]\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})$$

$$=J^2[|\psi(\mathbf{R}_1\mathbf{R}_2)|^2-|\psi(\mathbf{R}_1\mathbf{R}_1)|^2]$$

$$\times\frac{\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})}{(\omega+h_{1+}+h_{2-})^2-(h_{1+}-h_{2-})^2} \quad (5.1)$$

by simple algebraic transformations. We shall use again, as in I, a simple cubic lattice and the extreme tight-binding approximation:

$$T(k)=-2T\sum_{a=1}^{3}\cos k_aR. \quad (5.2)$$

Introducing the dimensionless variables

$$\phi_a=k_aR, \quad (5.3)$$

one may write

$$h_{1+}(\phi_1)+h_{2-}(\phi_2)=J+4T\sum_{a=1}^{3}\sin\left(\frac{\phi_{1a}+\phi_{2a}}{2}\right)$$

$$\times\sin\left(\frac{\phi_{1a}-\phi_{2a}}{2}\right), \quad (5.4)$$

$$h_{1+}(\phi_1)-h_{2-}(\phi_2)=-4T\sum_{a=1}^{3}\cos\left(\frac{\phi_{1a}+\phi_{2a}}{2}\right)$$

$$\times\cos\left(\frac{\phi_{1a}-\phi_{2a}}{2}\right). \quad (5.5)$$

The quantity called difference of momentum in I,

$$\Phi_a=\phi_{1a}-\phi_{2a}, \quad (5.6)$$

can be considered also as the center-of-mass momentum of the electron-hole pair. In I, it was found that the pair energy is smallest if $\Phi_a=\pi$ for all three of its components. This is then the only case we shall treat here, for which

$$h_{1+}(\phi_1)-h_{2-}(\phi_2)=0. \quad (5.7)$$

The solution of Eq. (5.1) is greatly simplified. It can be written in the form

$$(\omega+h_{1+}+h_{2-}-J\delta_{\mathbf{R}_1\mathbf{R}_2})\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})$$

$$=J^2[|\psi(\mathbf{R}_1\mathbf{R}_2)|^2-|\psi(\mathbf{R}_1\mathbf{R}_1)|^2]\frac{\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})}{(\omega+h_{1+}+h_{2-})}. \quad (5.8)$$

The left-hand side here is written in the form called "first formulation of the eigenvalue problem" in I while the right-hand side corresponds to the "second formulation."

Now, this last equation is valid in the insulating range in which we expect the electron-hole wave function to be reasonably well localized on one atom, i.e., we expect

$$\frac{|\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})|^2}{|\psi(\mathbf{R}_{1+}\mathbf{R}_{1-})|^2}\ll1; \quad 1\neq2. \quad (5.9)$$

Looking at the right-hand side of Eq. (5.8), we then see that the $|\psi(\mathbf{R}_1\mathbf{R}_2)|^2$ coefficient will only contribute a third-order term to the equation. We could then approximate

$$|\psi(\mathbf{R}_1\mathbf{R}_2)|^2-|\psi(\mathbf{R}_1\mathbf{R}_1)|^2$$

$$\approx|\psi(\mathbf{R}_1\mathbf{R}_1)|^2[\delta_{\mathbf{R}_1\mathbf{R}_2}-1] \quad (5.10)$$

in Eq. (5.7), and get

$$(\omega+h_{1+}+h_{2-}-J\delta_{\mathbf{R}_1\mathbf{R}_2})\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})$$

$$=J^2|\psi(\mathbf{R}_1\mathbf{R}_1)|^2[\delta_{\mathbf{R}_1\mathbf{R}_2}-1]\frac{\psi(\mathbf{R}_{1+}\mathbf{R}_{2-})}{(\omega+h_{1+}+h_{2-})}. \quad (5.11)$$

Hopefully, the solution to this last equation should verify condition (5.9). Without expanding again the details of the transformation between Wannier and Bloch representations we only set forth the final result:

$$\left(\Omega+1+\frac{c}{2}\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}}\right)\psi(\mathbf{k})-N^{-1}\sum_{\mathbf{k}'}\psi(\mathbf{k}')$$

$$=-|\psi(\mathbf{R}_1\mathbf{R}_1)|^2\left\{\frac{\psi(k)}{(\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}})}\right.$$

$$\left.-N^{-1}\sum_{\mathbf{l}}\frac{\psi(k')}{(\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}'\cdot\mathbf{l}})}\right\}, \quad (5.12)$$

where

$$\psi(\mathbf{R}_1\mathbf{R}_2)=N^{-1/2}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(\mathbf{R}_1-\mathbf{R}_2)}\psi(\mathbf{k}), \quad (5.13)$$

$$\Omega=\omega/J, \quad (5.14)$$

$$c=4T/J, \quad (5.15)$$

and where the summations $\mathbf{l}$ have to be extended over the six nearest neighbors. Using Eq. (5.12) itself to evaluate the summation terms which appear in it, we get the eigenvalue condition

$$\sum_{\mathbf{k}}\left\{\frac{|\psi(\mathbf{R}_1\mathbf{R}_1)|^2+\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}}}{(\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}})^2+|\psi(\mathbf{R}_1\mathbf{R}_1)|^2}\right\}=N, \quad (5.16)$$

which must be solved self-consistently with the requirement that

$$|\psi(\mathbf{R}_1\mathbf{R}_1)|^2=|\sum_{\mathbf{k}}\psi(\mathbf{k})|^2=|A|^2$$

$$\times\left|\sum_{\mathbf{k}}\left[\frac{\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}}}{(\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}})^2+|\psi(\mathbf{R}_1\mathbf{R}_1)|^2}\right]\right|^2, \quad (5.17)$$

where $|A|^2$ is the normalization constant determined by

$$\sum_{\mathbf{R}_2}|\psi(\mathbf{R}_1\mathbf{R}_2)|^2=|A|^2$$

$$\times\sum_{\mathbf{k}}\left|\frac{\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}}}{(\Omega+1+\frac{1}{2}c\sum_{\mathbf{l}} e^{i\mathbf{k}\cdot\mathbf{l}})^2+|\psi(\mathbf{R}_1\mathbf{R}_1)|^2}\right|^2=1. \quad (5.18)$$

The self-consistent solution of the set of equations (5.16)–(5.18) by a trial and error iteration procedure yields the eigenvalue curve of Fig. 1. At the cutoff point (c_M,Ω_M) the value of

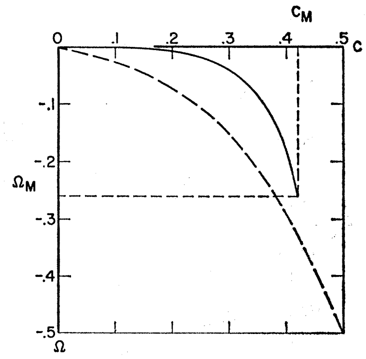

FIG. 1. Eigenvalue Ω as a function of c and their critical values. Solid line—self-consistent; dashed line—non-self-consistent.

$$\frac{|\psi(\mathbf{R}_1\mathbf{R}_2)|^2}{|\psi(\mathbf{R}_1\mathbf{R}_1)|^2}\lesssim 0.01 \quad (5.19)$$

was found to be at its maximum. This then justifies our assumption in Eq. (5.9).

Since the eigenvalue equation (4.20) was derived for the insulating range, it follows that the system under study is an insulator for $0\leq c\leq c_M$ but is not for $c>c_M$ since there is no longer a solution to (4.20) for those values. The Mott transition then occurs at a critical value of

$$c_M=0.42 \quad \text{or} \quad T/J=0.105. \quad (5.20)$$

Now, physically, one would expect the Mott transition to occur when the energy of the bound-electron–hole pair becomes equal to the energy of the free-electron–hole pair with the same center-of-mass momentum, i.e., when

$$w_{\text{pair}}/J=-3c=\text{free bandwidth}. \quad (5.21)$$

Looking at our effective Hamiltonian (2.5), we see that the energy of the pair is given by

$$w_{\text{pair}}=N^{-1}\left\langle H'-\frac{J}{2}\sum_i(n_{i+}+n_{i-})\right\rangle,$$

or

$$w_{\text{pair}}=\omega-\tfrac{1}{2}J(\langle n_{i+}\rangle+\langle n_{i-}\rangle),$$

or

$$w_{\text{pair}}/J=\Omega-1. \quad (5.22)$$

This normalized energy of the pair is plotted in Fig. 2.

It is immediately apparent that Eqs. (5.20) and (5.21) give the same result at $c=c_M$, as required.

The critical value of c in the non-self-consistent calculation, i.e., with the omission of the quadratic term in the potential in Eq. (3.5), turned out to be 0.5 in I. The decrease of the critical value was to be expected on physical grounds. In the non-self-consistent calculation the depth of the potential well and its width for a pair are independent of the motion of the other pairs. This means that the members of the selected pair, when they make excursions to neighboring atoms find exactly one electron and one hole on these atoms. In reality the

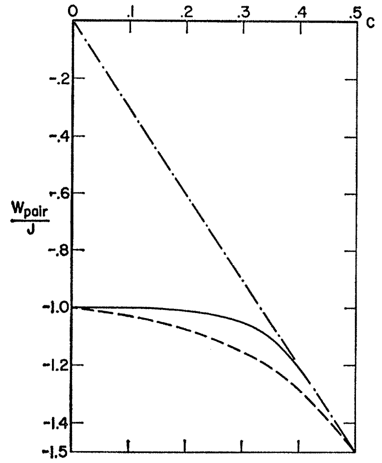

FIG. 2. Pair energy as a function of c. Solid line—self-consistent; dashed line—non-self-consistent; dotted line—free pair.

neighboring atoms are subject to the same dynamics and if the selected pair can make excursions, so can the others. Thus the selected pair is not certain to find a full-fledged pair on the neighboring sites and this makes the effective potential well shallower. The dissociation of the pair occurs at smaller band energies if this effect is taken into account.

Equation (5.19) shows that the pair is almost entirely concentrated on one atom. The self-consistent calculation requires better concentration for binding than the non-self-consistent one. So it appears that the better approximation yields better localization. One may conjecture that an exact solution would result in a pair completely localized on one atom.

The study of Fig. 2 suggests that the exact solution would yield $w_{\text{pair}}/J=-1$ with the critical value of $c_M=\frac{1}{3}$.

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Erratum

Exchange Model of Zero-Bias Tunneling Anomalies, JOEL A. APPELBAUM [Phys. Rev. **154**, 633 (1967)]. The captions associated with Figs. 7 and 8 should be interchanged.