PHYSICAL REVIEW B VOLUME 8, NUMBER 10 15 NOVEMBER 1973

Electron Correlation and Metal-Nonmetal Transition in a Disordered Binary System*†

Fumiko Yonezawa‡
Belfer Graduate School of Science, Yeshiva University, New York, New York 10033

Mitsuo Watabe
Department of Physics, Tohoku University, Japan

(Received 8 January 1973)

The metal-insulator transition due to electron correlation and potential fluctuations in a substitutionally disordered n-component system, particularly in a binary disordered system, is studied on the basis of the Hubbard model and of the localization-delocalization concept in the Anderson sense. For illustration, the case of $n = 1$ is first studied. This is the original Hubbard system of a regular crystal, for which the mobility gap coming from the random distribution of spins is calculated according to the localization function. The existence of the mobility gap alters not only the critical metal-nonmetal (M-NM) density but also the sharpness of the transition in the sense that the critical index of the mobility gap is 1/2, while that of the density-of-states gap is 3/2. It is shown that for reasonably general systems the resonance corrections in the Hubbard approximation modify the effect of the scattering corrections quantitatively rather than qualitatively. It is proved that, when only the scattering correction of the Hubbard theory is taken into account, the problem of treating the effect of electron correlation in an n-component alloy with substitutional disorder is reduced to that of treating an independent-electron picture in the coherent-potential approximation for a $2n$-component system. For binary systems with $n = 2$, the quasiparticle densities of states are evaluated for various interatomic distances and the localization of the states is examined. Numerical results are discussed with emphasis on the M-NM transition. A possible extension of the method to amorphous systems with topological disorder is mentioned in relation to experiments on the M-NM transition in some metal rare-gas solids.

I. INTRODUCTION

In connection with the problem of localization of electrons in certain random lattices,[1] the metal-nonmetal (M-NM) transition in disordered systems has recently aroused increasing attention.[2,3] The nature of the transition in disordered systems is generally multifold even when we confine ourselves to such a case in which the system does not undergo a change in the atomic structure at the transition point. Experimentally, the M-NM transition has been observed in doped semiconductors, metal-ammonia solutions,[4] mixed crystals or mixed noncrystalline solids composed of both metallic and nonmetallic elements,[5-7] and fluid metals under supercritical conditions.[8,9] The metal-insulator transitions in these systems are usually induced by changing the effective or averaged interatomic distance somehow, and this change of the effective interatomic distance over a wide range of values is attained at the expense of a regular array of atoms, which means that the atomic configurations of these systems are inevitably made disordered by doping, mixing, alloying, or applying a supercritical condition. As a result, potential fluctuations do exist in these systems and the localization mechanism of electrons due to the potential fluctuations in the Anderson sense[1] sets in. Out of the other possible mechanisms for the transition, the Coulomb interaction between electrons is considered to be dominant especially for a system composed of open-shell metallic atoms. This type of M-NM transition has first been suggested by Mott[10] for a regular array of hydrogen atoms and is called the Mott transition. In order to treat the effect of electron correlations in a regular system, Hubbard[11] has introduced a model which has been successful to some extent in explaining the Mott transition.

The purposes of this paper are to study the effect of electron correlation in substitutionally disordered systems and particularly to see how the M-NM transitions in these systems are described in an interacting-electron picture. We employ the Hubbard model for electron correlation. The effect of alloying is formulated on the basis of the coherent-potential approximation (CPA).[12] First, we discuss the mobility gap[13,14] in the Hubbard band. Our special interest is centered around the case where the array of atoms is regular. It is shown that even in this case the states at the extreme edges of the quasiparticle density of states are localized because of the random configuration of spins.[15] The disappearance of the mobility gap rather than that of the density-of-states gap yields the transition from an insulator to a metal. The way in which the mobility gap approaches zero is discussed in relation to the sharpness of the transition. We mention that it is enough to take into account only the scattering corrections when the qualitative behaviors of electrons are required and that the Hubbard model applied to a disordered binary alloy is reduced to the problem of solving equations for a site-random alloy made of four components in an independent-particle scheme and more generally, the Hubbard model in an n-component alloy[16] is identical with the CPA for a $2n$-component alloy.

In the next place, we show the results of a numerical calculation of the density of states and the mobility edges obtained from the above-described formulations for substitutionally disordered binary alloys and discuss the effect of potential fluctuations. Finally, some comments are made on the possible application of the present theory to the problem of the M-NM transition in some metal-rare-gas systems.[5-7]

II. MOBILITY GAP IN HUBBARD BAND

For later convenience, in this section we study the original Hubbard system which corresponds to a regular array of atoms. The system is described by the Hamiltonian

$$H=\sum_{i,\sigma} T_0 n_{i\sigma}+\sum_{i\neq j}\sum_{\sigma} t_{ij} a^{\dagger}_{i\sigma} a_{j\sigma}+\sum_{i,\sigma}\frac{U}{2} n_{i\sigma} n_{i-\sigma} , \tag{2.1}$$

in which $a^{\dagger}_{i\sigma}$ and $a_{i\sigma}$ are, respectively, the creation and destruction operators for an electron of spin σ in the atomic orbital on the ith site and $n_{i\sigma}$ is the corresponding number operator; T_0 is the binding energy of the atomic level and U is the Coulomb interaction of two electrons with opposite spins on the same site; t_{ij} designates the transfer matrix between site i and j. It is assumed that there is one atomic orbital per site. We shall be concerned with a nonmagnetic system with a half-filled s band. It has been concluded that, when a generalized theory developed by Hubbard is applied to a special case of unperturbed band structure with the density of states of parabolic form

$$\begin{aligned}\rho_0(E)&=\frac{4}{\pi\Delta}\left[1-\left(\frac{E-T_0}{\frac{1}{2}\Delta}\right)^2\right]^{1/2} \quad &&\text{if } |E-T_0|<\tfrac{1}{2}\Delta\\ &=0 &&\text{otherwise} ,\end{aligned} \tag{2.2}$$

where Δ is a bandwidth, there exists a critical ratio $(\Delta/U)_c$ at which a qualitative change of electronic structure takes place. The situation is illustrated in Fig. 1(a) where the density of states is nonvanishing in the hatched region of the $(E/U, \Delta/U)$ plane, while those states indicated by the double hatching are occupied at $T=0$. More precisely, the quasiparticle spectrum consists of two bands when the ratio Δ/U is smaller than this critical value while it consists of a single band when Δ/U exceeds $(\Delta/U)_c$, thus lending support to the transition from an insulator to a conductor as suggested by Mott. Note that, in the above analysis, both the scattering and resonance corrections are taken into account.

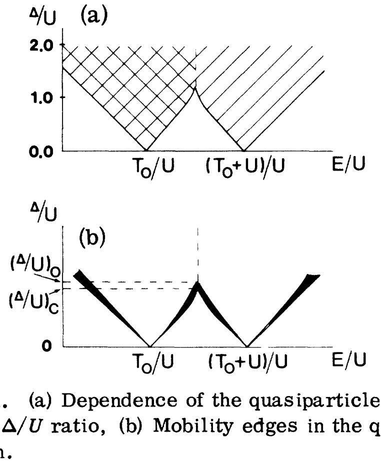

FIG. 1. (a) Dependence of the quasiparticle spectrum upon the Δ/U ratio, (b) Mobility edges in the quasiparticle spectrum.

It is interesting to mention that the density of states in Fig. 1(a) is that for quasiparticles and thus each state has a finite lifetime. This comes from the fact that electronic states are modified by scattering and resonance corrections since the distribution of spins are random in the Hubbard model in spite of a regular configuration of atoms. In view of the fact that the random configuration of spins in an interacting-electron model is regarded to be analogous to some extent to the fluctuations of atomic potential for an independent electron, the states near the band edges of the Hubbard band are expected to be localized in the Anderson sense as schematically illustrated in Fig. 1(b). The possibility of localization of states at the extreme edges of the quasiparticle density of states has been pointed out.[17,18] We show in the following that this is actually the case.[15]

A. Density-of-States Gap

First, let us consider a special nonmagnetic system with a half-filled s band of structure described by Eq. (2.2) and show that the density-of-states gap near the critical ratio $(\Delta/U)_c=\delta_c$ approaches zero as[11]

$$E_g=(\text{const})[(\Delta/U)_c-(\Delta/U)]^{3/2} . \tag{2.3}$$

The equation for the self-energy Σ in the Hubbard approximation is written[11]

$$E\Sigma^3-(\tfrac{2}{3}E^2+\tfrac{1}{4}U^2-\tfrac{3}{16}\Delta^2)\Sigma^2+\tfrac{1}{12}EU^2\Sigma+\tfrac{1}{48}U^4=0 , \tag{2.4a}$$

or with dimensionless variables normalized by U, $\epsilon=E/U$, $\zeta=\Sigma/U$, and $\delta=\Delta/U$, we have

$$\epsilon\zeta^3-(\tfrac{2}{3}\epsilon^2+\tfrac{1}{4}-\tfrac{3}{16}\delta^2)\zeta^2+\tfrac{1}{12}\epsilon\zeta+\tfrac{1}{48}=0 , \tag{2.4b}$$

where the origin of energy is chosen such that $T_0+\frac{1}{2}U=0$.

The density of states is nonzero when the solutions of Eq. (2.4b) have nonzero imaginary parts. The whole formulation is symmetric with respect to $\epsilon=0$. Remembering that in the case under consideration the Fermi level is at $\epsilon=0$, we can obtain the critical ratio by equating ϵ to zero in Eq. (2.4b) and searching the condition that ζ has nonreal solutions, i.e.,

$$3(4-3\delta^2)=(\zeta^{-1})^2<0 , \tag{2.5}$$

which yields the critical ratio $\delta_c = (\Delta/U)_c = (\frac{4}{3})^{1/2}$. By expanding ϵ, ζ^{-1}, and δ in the vicinity of the critical values, namely, at $\epsilon = \zeta^{-1} = 0$ and $\delta = \delta_c$, and keeping with lowest orders in ϵ and $\delta_c - \delta$, we have $\epsilon = \text{const}(\delta_c - \delta)^{3/2}$ which gives Eq. (2.3).

B. Localization of Electrons at Extreme Edges of the Hubbard Band

Next, we examine whether or not the states at the extreme edges in the bands shown in Fig. 1(a) are localized as indicated in Fig. 1(b). For testing the localization of states, we employ the criterion introduced by Economou and Cohen.[19] According to their argument, the states corresponding to energy E are localized if the localization function $F(E)$ defined by

$$F(E) = \frac{\frac{1}{2}\Delta}{|E - \Sigma|} = \frac{\frac{1}{2}\delta}{|\epsilon - \zeta|} \tag{2.6}$$

is smaller than 1.

First, let us consider the case $\epsilon = 0$ and work out the regions of δ for which the density of states are nonzero but the states are localized. From Eq. (2.5), it is shown that the density of states is nonvanishing when

$$\delta > \delta_c = (\tfrac{4}{3})^{1/2} = 1.15\ldots\,. \tag{2.7}$$

On the other hand, for δ satisfying Eq. (2.7), the condition for localization is given by Eq. (2.6) as

$$F(\epsilon = 0) = \tfrac{1}{2}\delta\,|\,\zeta^{-1}\,| = \tfrac{1}{2}\delta[3(3d^2 - 4)]^{1/2} < 1\ , \tag{2.8a}$$

which leads to an inequality

$$\delta < [\tfrac{2}{3}(1 + \sqrt{2})]^{1/2} = 1.27\ldots\,. \tag{2.8b}$$

As a result, the required region for Δ is expressed as

$$(\tfrac{4}{3})^{1/2} < \delta < [\tfrac{2}{3}(1 + \sqrt{2})]^{1/2}\ . \tag{2.9}$$

Let us denote the upper bound of region (2.9) by $(\Delta/U)_0 = \delta_0$. On inspection of Fig. 1(b), this ratio corresponds to a critical ratio at which the mobility gap disappears.

Secondly, let us investigate the properties of the states for a fixed value of δ at $\delta_c = (\frac{4}{3})^{1/2}$. For this critical ratio, Eq. (2.4b) is reduced to

$$\epsilon\zeta^3 - \tfrac{2}{3}\epsilon^2\zeta^2 + \tfrac{1}{12}\epsilon\zeta + \tfrac{1}{48} = 0\ . \tag{2.10}$$

It is easy to derive from this equation that, for small values of ϵ, one real solution of $\zeta = \zeta_R$ is expanded as follows:

$$\zeta_R = \frac{-1}{2(6)^{1/3}}\ \frac{1}{e}\ [1 - (\tfrac{4}{81})^{1/3}e^2 + \cdots]\ , \tag{2.11}$$

where $e = \epsilon^{1/3}$. In terms of ζ_R, Eq. (2.10) is arranged in the following way:

$$\epsilon(\zeta - \zeta_R)(\zeta^2 + \zeta_R\zeta + \zeta_R^2 + \tfrac{1}{12}) = 0\ , \tag{2.12}$$

from which we have the other two complex-conjugate solutions in the form

$$\zeta = \tfrac{1}{2} - \zeta_R \pm \tfrac{1}{2}i(3\zeta_R^2 + \tfrac{1}{3})^{1/2}\ . \tag{2.13}$$

Keeping with the lowest order of e, we obtain the criterion for localization as

$$F(\,|\,\epsilon\,| \ll 1) = \tfrac{1}{2}\delta_c\,|\,\epsilon - \zeta\,|^{-1} = \tfrac{1}{2}\delta_c\,|\,\zeta_R\,|^{-1} < 1\ , \tag{2.14}$$

which yields

$$|\,\epsilon\,| < \tfrac{1}{16}\sqrt{3} = 0.108\ldots\,. \tag{2.15}$$

Thus, the states corresponding to the energy region given by Eq. (2.15) and to the critical ratio δ_c are localized. Based upon the above discussion, we can draw a picture as shown in Fig. 2 in the immediate vicinity of the point $(0, \delta_c)$ in the $(E/U, \Delta/U)$ plane. As in Fig. 1, the hatched region denotes that the density of states is not zero; the double hatching represents the occupied states at $T = 0$ and the shadowed region indicates that the corresponding states are localized.

C. Mobility Gap in the Hubbard Bands

In view of the fact that the band is half-filled and therefore the Fermi level is at $\epsilon = 0$ as shown in Fig. 2, it follows directly that at $T = 0$ the system remains nonconductive even after Δ/U exceeds $(\Delta/U)_c$ since the eigenstates at the Fermi level are in the mobility gap and as a result are still localized. Therefore, the critical value Δ/U for the metal-nonmetal transition should be determined by the point $(\Delta/U)_0$ at which the mobility gap disappears rather than by the point $(\Delta/U)_c$ at which the density-of-states gap is filled. Furthermore, the activation energy E_g for electric conduction at $T \neq 0$ should be described by the mobility gap E_{mg} rather than the density-of-states gap E_{dg}.

The mobility gap E_{mg} calculated according to the criterion $F(E) = 1$ is illustrated in Fig. 3 in comparison with the density-of-states gap E_{dg}.

It is also interesting to see how the mobility gap

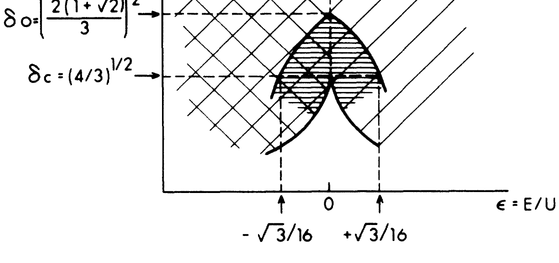

FIG. 2. Density of states and the mobility edges near the critical ratios $(\Delta/U)_c = \delta_c$ and $(\Delta/U)_0 = \delta_0$ in the $(E/U, \Delta/U)$ plane.

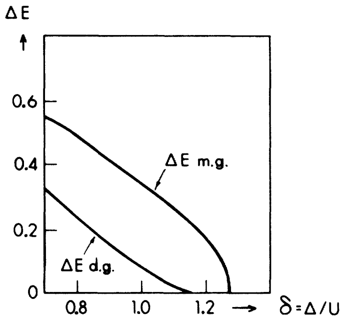

FIG. 3. Dependence of the mobility gap E_{mg} and the density-of-states gap E_{dg} upon the Δ/U ratio near the critical values. (Both the scattering and resonance corrections are taken into account).

between two succeeding mobility edges becomes zero as $\delta = \Delta/U$ approaches from below the critical value $(\Delta/U)_0$ of the mobility gap. To this end, let us expand ζ around $\zeta = \zeta_0$ which is the solution of Eq. (2.4b) for $\epsilon = 0$ and $\delta = \delta_0$, and is given by

$$-\left(\tfrac{1}{4} - \tfrac{3}{16}\,\delta_0^2\right)\zeta_0^2 + \tfrac{1}{48} = 0 \tag{2.16a}$$

or by

$$\zeta_0 = i\left[\tfrac{1}{6}(1+\sqrt{2})\right]^{1/2} \equiv i\alpha_0 \ . \tag{2.16b}$$

With $\zeta = \zeta_0 + \zeta_1$ and $\delta = \delta_0 - \delta_1$, Eq. (2.4b) is rewritten

$$\epsilon(\zeta_0^3 + 3\zeta_0^2\zeta_1) - \left[\tfrac{2}{3}\epsilon^2 + \tfrac{1}{4} - \tfrac{3}{16}(\delta_0^2 - 2\delta_0\delta_1)\right] \times (\zeta_0^2 + 2\zeta_0\zeta_1) + \tfrac{1}{12}\epsilon(\zeta_0 + \zeta_1) + \tfrac{1}{48} = 0 \ . \tag{2.17}$$

Then we have, for ζ_1,

$$\zeta_1 = \left(\frac{2\sqrt{2}+1}{12}\right)\left(\frac{\sqrt{2}+1}{6}\right)^{1/2}\epsilon + i\left(\frac{\sqrt{2}+1}{6}\right) \times \left[\tfrac{2}{3}\epsilon^2 + \frac{3}{8}\left(\frac{2+2\sqrt{2}}{3}\right)^{1/2}\delta_1\right] 2\left(\frac{\sqrt{2}+1}{6}\right)^{1/2} \times \left[\frac{\sqrt{2}-1}{8} - \tfrac{2}{3}\epsilon^2 - \frac{3}{8}\left(\frac{2+2\sqrt{2}}{3}\right)^{1/2}\delta_1\right] + i\left(\frac{6\sqrt{2}+5}{12}\right)\epsilon \ . \tag{2.18}$$

The criterion for the mobility edge now becomes

$$F(\epsilon) = \frac{\tfrac{1}{2}(\delta_0 - \delta_1)}{|\epsilon - \zeta_0 - \zeta_1|} = 1 \ , \tag{2.19a}$$

or equivalently

$$\epsilon^2 - 2\epsilon\alpha_1 + \alpha_1^2 + 2\alpha_0\alpha_2 + \alpha_2^2 + \tfrac{1}{2}\delta_0\delta_1 = 0 \ , \tag{2.19b}$$

where we write $\zeta_1 \equiv \alpha_1 + i\alpha_2$ and Eq. (2.16) has been made use of. From Eq. (2.18), it is easily shown that $\alpha_1 = O(\epsilon)$ and $\alpha_2 = O(\epsilon^2) + O(\delta_1)$, which together with Eq. (2.19b) leads to the relation $\epsilon^2 \propto \delta_1$ or $\epsilon \propto (\delta_1)^{1/2}$. Thus, the mobility gap E_{mg} goes to zero at $\Delta/U \to (\Delta/U)_0$ as

$$E_{mg} = (\text{const})[(\Delta/U)_0 - (\Delta/U)]^{1/2} \ . \tag{2.20}$$

Consequently, it is concluded that the critical behavior of E_{mg} qualitatively differs from the behavior of E_{dg} in the sense that the derivative of E_{mg} at $(\Delta/U)_0$ is divergent while that of E_{dg} at $(\Delta/U)_c$ is zero as is apparent from Eq. (2.3). This fact is interesting in relation with the problem about the sharpness of the Mott transition and with the interpretation of the activation energy ϵ_2 in the problem of impurity conduction.[20]

D. Discussion

As mentioned at the beginning of this section, all the formulations in this section have been carried out on the basis of the original Hubbard theory in which both the scattering and resonance (or dynamical) corrections are taken into account. However, it is easy to see that the resonance correction terms give only quantitative modifications to the scattering corrections when the system is assumed to be nonmagnetic and the density-of-states function of a parabolic form as given by Eq. (2.2) is employed for the unperturbed band structure. When only the scattering corrections are included in the theory, a self-consistent equation for a self-energy Σ is, instead of Eqs. (2.4a) and (2.4b), written

$$E\Sigma^3 - \tfrac{1}{4}(U^2 - \tfrac{1}{4}\Delta^2)\Sigma^2 - \tfrac{1}{4}EU^2\Sigma + \tfrac{1}{16}U^4 = 0 \ , \tag{2.21a}$$

or as

$$\epsilon\zeta^3 - \tfrac{1}{4}(1 - \tfrac{1}{4}\delta^2)\zeta^2 - \tfrac{1}{4}\epsilon\zeta + \tfrac{1}{16} = 0 \ , \tag{2.21b}$$

where the symbols have the same meanings as before. From Eq. (2.21b), it is readily shown after some simple manipulation that the critical index of the density-of-states gap at the critical point $(\Delta/U)_0$ is $\frac{3}{2}$ while that of the mobility gap at $(\Delta/U)_0$ is $\frac{1}{2}$ just as in the case of the original Hubbard approximation discussed in this section.

The mobility gap and the density-of-states gap in the present example are shown in Fig. 4. On comparing Figs. 3 and 4, it is apparent that the resonance correction does not introduce any qual-

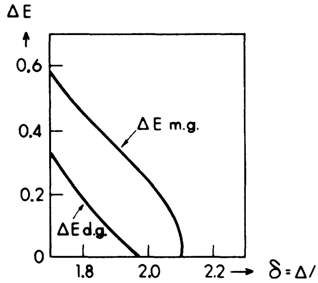

FIG. 4. Dependence of the mobility gap E_{mg} and the density-of-states gap E_{dg} upon the Δ/U ratio near the critical values. (Only the scattering corrections are taken into account.)

itative alteration; therefore for getting qualitative properties of electrons, it is reasonable to include only the scattering effects.

III. HUBBARD MODEL FOR A DISORDERED n-COMPONENT ALLOY

The Hubbard model for a binary alloy has been discussed by Fukuyama and Ehrenreich.[21] We extend the theory to a more general case of an n-component alloy and show that the problem is reduced to that of finding a self-consistent solution of the CPA in a $2n$-component alloy.

We work with the Hamiltonian of the form

$$H=\sum_{i,\sigma} E_i a_{i,\sigma}^{\dagger} a_{i,\sigma}+\sum_{i\neq j}\sum_{\sigma} t_{ij} a_{i,\sigma}^{\dagger} a_{j,\sigma} + \sum_{i,\sigma}\frac{1}{2} U_i n_{i,\sigma} n_{i,-\sigma} \qquad (3.1)$$

where the notation is the same as for Eq. (2.1) except that E_i is an atomic level of an atom at site i and U_i is the Coulomb interaction between electrons with opposite spins on the same site i. Let us consider an n-component alloy and assume that the atomic distribution is completely random. We assume that E_i takes atomic levels $E_1, E_2, \ldots, E_s, \ldots, E_n$ with respective probabilities $x_1, x_2, \ldots, x_s, \ldots, x_n$, and U_i equals one of n values $U_1, U_2, \ldots, U_s, \ldots, U_n$. The off-diagonal matrix element t_{ij} is supposed to be independent of the types of atoms at i and j.

For illustration, we shall be concerned in the present section only with the scattering corrections since, as has been shown in the preceding section, in moderately general systems the resonance or dynamical corrections give rise to quantitative modifications rather than qualitative ones.

It is known that the scattering corrections of the Hubbard theory are described in the alloy analogy and thus determined from the relation equivalent to the CPA formulation. Therefore, the effective atomic energy $\Sigma_{s,\sigma}$ for an electron with spin σ at a site of sth component atom is given by

$$\frac{n_{s,-\sigma}(E_s+U_s-\Sigma_{s,\sigma})}{1-(E_s+U_s-\Sigma_{s,\sigma})\,G_{s,\sigma}}+\frac{(1-n_{s,-\sigma})(E_s-\Sigma_{s,\sigma})}{1-(E_s-\Sigma_{s,\sigma})\,G_{s,\sigma}}=0\ , \qquad (3.2)$$

where the partial occupation number $n_{s,\sigma}$ on the sth component atom is related to the partial Green function $G_{s,\sigma}$ by

$$n_{s,\sigma}=-\pi^{-1}\,\mathrm{Im}\int_{-\infty}^{E_F} G_{s,\sigma}\,dE\ . \qquad (3.3a)$$

Note that the total occupation number n_σ is given by

$$n_\sigma=\sum_s x_s n_{s,\sigma}\ . \qquad (3.3b)$$

Here E_F is the Fermi energy which is not given *a priori* but must be determined self-consistently. The partial Green's function $G_{s,\sigma}$ describes the effect of replacing one atom by an impurity atom having an atomic level $\Sigma_{s,\sigma}$ in an otherwise regular crystal. Recalling the formulation for the repeated scattering due to a single impurity, we can express $G_{s,\sigma}$ in terms of the Green's function G_σ and the effective atomic level Σ_σ for the regular system as

$$G_{s,\sigma}=G_\sigma+G_\sigma(\Sigma_{s,\sigma}-\Sigma_\sigma)\,G_{s,\sigma} \qquad (3.4a)$$

or

$$G_{s,\sigma}=\frac{1}{G_\sigma^{-1}-(\Sigma_{s,\sigma}-\Sigma_\sigma)}\ . \qquad (3.4b)$$

On the other hand, the regular system defined by G_σ and Σ_σ is taken to be an effective averaged crystal in the sense that both the random configurations of spins and the random distributions of atoms are averaged over, and accordingly the system has restored the translationally invariant symmetry. Since the average over the spin configuration is effected by Eq. (3.2) in the alloy analogy, the remaining process for obtaining G_σ and Σ_σ is the average over the atomic configurations. This is implemented as follows. The site-diagonal element of the ensemble-averaged Green's function G_σ is expressed by the Fourier component $G_{\vec{k}}(E)$ while $G_{\vec{k}}(E)$ is defined by means of the total self-energy or the coherent potential Σ_σ as

$$G_\sigma(E)=\frac{1}{N}\sum_{\vec{k}} G_{\vec{k}}(E)=\frac{1}{N}\sum_{\vec{k}}\frac{1}{E-E_{\vec{k}}-\Sigma_\sigma}\ , \qquad (3.5)$$

where $E_{\vec{k}}$ is the band energy determined from the transfer matrix t_{ij}. This self-energy Σ_σ is in turn related to the total Green's function by the CPA equation

$$\left\langle\frac{\Sigma_{i,\sigma}-\Sigma_\sigma}{1-(\Sigma_{i,\sigma}-\Sigma_\sigma)\,G_\sigma}\right\rangle=\sum_{s=1}^{n}\frac{x_s(\Sigma_{s,\sigma}-\Sigma_\sigma)}{1-(\Sigma_{s,\sigma}-\Sigma_\sigma)\,G_\sigma}=0\ , \qquad (3.6)$$

where the angular brackets represent the average over the atomic configurations. By definition, the partial Green's function for an sth component atom is connected to the total Green's function by

$$G_\sigma=\langle G_{i,\sigma}\rangle=\sum_{s=1}^{n} x_s G_{s,\sigma}\ , \qquad (3.7a)$$

which, together with Eqs. (3.3a) and (3.3b), naturally leads to

$$n_\sigma=-\pi^{-1}\,\mathrm{Im}\int_{-\infty}^{E_F} G_\sigma(E)\,dE\ . \qquad (3.7b)$$

Equations (3.2)–(3.7) and analogous equations for an electron with spin $-\sigma$ should be solved simultaneously, and the solution gives the properties of interacting electrons in an n-component alloy with site-diagonal randomness. (A detailed procedure for attaining the self-consistency in actual calculations will be shown in a succeeding section.)

Now, let us show that the problem of finding the solution of the above simultaneous equations is reduced to that of solving the CPA equation for a $2n$-component alloy. It has been shown by Yonezawa[16] that the CPA equation for a disordered n-component alloy is rearranged into a simpler form on the introduction of a new variable. For illustration, we employ a one-electron Hamiltonian of the following form

$$H=\sum_i E_i a_i^\dagger a_i + \sum_{i,j} t_{ij} a_i^\dagger a_j \ . \tag{3.8}$$

This is the special case of Hamiltonian (3.1) in which all U_i's are zero. Here, the subscripts denoting the orientations of spins are dropped since interactions between electrons are not taken into account. Again, it is assumed that the Hamiltonian (3.8) bears site-diagonal disorder such that E_i takes n atomic levels $E_1, E_2, \ldots, E_s, \ldots, E_n$ with respective probabilities $x_1, x_2, \ldots, x_s, \ldots, x_n$. The CPA theory yields a well-known equation for an ensemble-averaged Green's function G and a coherent potential Σ as

$$\left\langle \frac{E_i-\Sigma}{1-(E_i-\Sigma)G} \right\rangle = \sum_{s=1}^{n} \frac{x_s(E_s-\Sigma)}{1-(E_s-\Sigma)G} = 0 \ . \tag{3.9}$$

It is shown that after some manipulation, Eq. (3.9) is brought to the CPA equation of a simpler form as

$$G=\left\langle \frac{1}{\xi-E_i} \right\rangle = \sum_{s=1}^{n} \frac{x_s}{\xi-E_s} \equiv \frac{1}{\xi-\Sigma} \ , \tag{3.10}$$

where

$$\xi \equiv G^{-1}+\Sigma \ . \tag{3.11}$$

Clearly, an equation analogous to Eq. (3.5) holds for G and Σ Along this line, Eq. (3.2) for the alloy analogy of the Hubbard model is rewritten as

$$G_{s,\sigma}=\frac{n_{s,-\sigma}}{\xi_{s,\sigma}-(E_s+U_s)}+\frac{1-n_{s,-\sigma}}{\xi_{s,\sigma}-E_s} \ , \tag{3.12}$$

in which

$$\xi_{s,\sigma}=G_{s,\sigma}^{-1}+\Sigma_{s,\sigma} \ . \tag{3.13}$$

In a completely similar way, Eq. (3.6) is rearranged into the following CPA equation:

$$G_\sigma=\left\langle \frac{1}{\xi_\sigma-\Sigma_{i,\sigma}} \right\rangle = \sum_{s=1}^{n} \frac{x_s}{\xi_\sigma-\Sigma_{s,\sigma}} \ , \tag{3.14}$$

with

$$\xi_\sigma \equiv G_\sigma^{-1}+\Sigma_\sigma \ . \tag{3.15}$$

It is easy to see that the relation

$$\xi_\sigma=\xi_{s,\sigma} \tag{3.16}$$

is derived from Eq. (3.4b). Substituting Eq. (3.16) into (3.12) and using the relation (3.7), we finally have

$$G_\sigma=\sum_{s=1}^{n}\left(\frac{x_s n_{s,-\sigma}}{\xi_\sigma-(E_s+U_s)}+\frac{x_s(1-n_{s,-\sigma})}{\xi_\sigma-E_s}\right) \ . \tag{3.17}$$

From the general discussion through Eqs. (3.8) to (3.11b), it is readily concluded that the simultaneous equations (3.5), (3.15), and (3.17) are identical with the CPA equation for a $2n$-component alloy with site-diagonal randomness in which an atomic level at an arbitrary site takes one of $2n$ values $E_1, E_2, \ldots, E_s, \ldots, E_n$ and $E_1+U_1, E_2+U_2, \ldots, E_s+U_s, \ldots, E_n+U_n$ with respective probabilities $x_1(1-n_{1,-\sigma}), x_2(1-n_{2,-\sigma}), \ldots, x_s(1-n_{s,-\sigma}), \ldots, x_n(1-n_{n,-\sigma})$ and $x_1 n_{1,-\sigma}, x_2 n_{2,-\sigma}, \ldots, x_s n_{s,-\sigma}, \ldots, x_n n_{n,-\sigma}$.

The only difference between the original CPA for a $2n$-component alloy and the Hubbard model for an n-component alloy is that in the latter problem the partial occupation numbers $n_{1,\sigma}, n_{2,\sigma}, \ldots$ must be determined in a self-consistent manner so as to satisfy a set of simultaneous equations for $n_{s,\sigma}$, E_F, Σ_σ, G_σ, etc., while in the former problem the concentration of each constituent atom is predetermined.

IV. METAL-NONMETAL TRANSITIONS IN DISORDERED BINARY ALLOYS

On account of the fact that the M-NM transition is experimentally observed in some binary systems,[5–7] let us particularly consider in the present section the case of disordered binary alloys. This example serves as an important model from which to learn essential aspects of the effect of electron correlations in systems with cellular disorder.[22] We can get as well some insight into the problem of the M-NM transition in some topologically disordered systems, which will be discussed in a succeeding section.

We formulate the problem based upon the general theory developed in preceding sections, calculate the density of states and the mobility edges, and discuss the effects of potential fluctuations on the M-NM transition. We study a disordered $A_x B_{1-x}$ alloy. In the Hamiltonian (3.1), the atomic level E_i is either E_A or E_B according as the site i is occupied by an atom A or B, respectively. Suppose E_A is smaller than E_B. The Coulomb energy U_i is also U_A or U_B for an A or B site, respectively. For numerical calculations we confine ourselves to the model with $U_B=0$ since this simulates a system where the effect of electron correlations in the lower band is important and the existence of B atoms serves as scattering centers, which is quite often the case in physically interesting systems. For instance, the concept of the present model is applicable to a doped semiconductor in which the activation energy ϵ_2 for an intermediate density region is regarded as originating from the Hubbard gap in the impurity band.[20] In a doped semiconductor, important in the study of the Mott transition, the existence of a conduction band should not be neglected even though the conduction band is rather far above the impurity band compared with the width of the impurity band.

For the band structure of the unperturbed crystal, we employ the parabolic form given by Eq. (2.2). Combining Eqs. (3.5) and (2.2), we have

$$G_\sigma(E)=\frac{1}{N}\sum_k \frac{1}{E-E_k-\Sigma_\sigma}=\int\frac{\rho_0(E')dE'}{E-E'-\Sigma_\sigma}\ . \tag{4.1}$$

The integration is performed with the result

$$G_\sigma(E)=\frac{8}{\Delta^2}\{E-\Sigma_\sigma-[(E-\Sigma_\sigma)^2-(\tfrac{1}{2}\Delta)^2]^{1/2}\}\ . \tag{4.2}$$

This is the very relation which leads to the self-consistent equations (2.4a) and (2.4b) [or to the equations (2.21a) and (2.21b)]. Straightforward algebra yields that, for this special model, ξ_σ defined by Eq. (3.15) is related to G_σ in the following way:

$$\xi_\sigma=G_\sigma^{-1}+\Sigma_\sigma=E-\tfrac{1}{16}\Delta^2G_\sigma\ . \tag{4.3}$$

Equations (3.17) and (4.3) applied to our system lead to the relation

$$E=\xi_\uparrow+\frac{\Delta^2}{16}\left(\frac{xn_{A\downarrow}}{\xi_\uparrow-(E_A+U_A)}+\frac{x(1-n_{A\downarrow})}{\xi_\uparrow-E_A}+\frac{1-x}{\xi_\uparrow-E_B}\right)\ , \tag{4.4a}$$

$$E=\xi_\downarrow+\frac{\Delta^2}{16}\left(\frac{xn_{A\uparrow}}{\xi_\downarrow-(E_A+U_A)}+\frac{x(1-n_{A\uparrow})}{\xi_\downarrow-E_A}+\frac{1-x}{\xi_\downarrow-E_B}\right)\ , \tag{4.4b}$$

where the partial occupation numbers $n_{A\uparrow}$ and $n_{A\downarrow}$ are related to the partial Green's functions $G_{A\uparrow}(E)$ and $G_{A\downarrow}(E)$ by Eq. (3.3a). Namely,

$$n_{A\uparrow}=-\pi^{-1}\int_{-\infty}^{E_F}\mathrm{Im}\,G_{A\uparrow}\,dE=-\pi^{-1}\times\int_{-\infty}^{E_F}\mathrm{Im}\left(\frac{n_{A\downarrow}}{\xi_\uparrow-(E_A+U_A)}+\frac{1-n_{A\downarrow}}{\xi_\uparrow-E_A}\right)dE\ , \tag{4.5a}$$

$$n_{A\downarrow}=-\pi^{-1}\int_{-\infty}^{E_F}\mathrm{Im}\,G_{A\downarrow}\,dE=-\pi^{-1}\times\int_{-\infty}^{E_F}\mathrm{Im}\left(\frac{n_{A\uparrow}}{\xi_\downarrow-(E_A+U_A)}+\frac{1-n_{A\uparrow}}{\xi_\downarrow-E_A}\right)dE\ , \tag{4.5b}$$

where use has been made of Eqs. (3.12) and (3.16). The Fermi energy which appears in Eqs. (4.5a) and (4.5b) is determined self-consistently so that the following equation for the averaged total occupation numbers is satisfied:

$$\begin{aligned}n_\sigma&=-\pi^{-1}\int_{-\infty}^{E_F}\mathrm{Im}\,G_\sigma(E)\,dE\\&=-\pi^{-1}\int_{-\infty}^{E_F}\mathrm{Im}(16/\Delta^2)(E-\xi_\sigma)\,dE\ .\end{aligned} \tag{4.6}$$

When the system is assumed to be nonmagnetic and the number of electrons present is one per A atom, then we have $n_\uparrow=n_\downarrow=\frac{1}{2}x$. The density of states is determined from $G_\sigma(E)$ by solving a set of simultaneous equations (4.4a)–(4.6). The actual implementation of a numerical calculation has been performed according to the following procedure: (i) give a test value of $n_{A\uparrow}(0\leq n_{A\uparrow}\leq\frac{1}{2})$; (ii) calculate $\xi_\downarrow$ by Eq. (4.4b) by using $n_{A\uparrow}$ given in (i); (iii) find E_F so that Eq. (4.6) is satisfied for $\xi_\downarrow$ derived in (ii) and $n_\downarrow=\frac{1}{2}x$; (iv) determine $n_{A\downarrow}$ according to Eq. (4.5b) on making use of $n_{A\uparrow}$ given in (i), $\xi_\downarrow$ calculated in (ii), and E_F defined in (iii); (v) solve Eq. (4.4a) for $\xi_\uparrow$ with $n_{A\downarrow}$ calculated in (iv); (vi) derive $n'_{A\uparrow}$ by Eq. (4.5a) with $n_{A\downarrow}$ and $\xi_\uparrow$ found in (iv) and (v), respectively, and with E_F found in (iii); and (vii) check whether $n'_{A\uparrow}$ obtained in (vi) is the same as $n_{A\uparrow}$ given in (i). The whole procedure is repeated with different test values for $n_{A\uparrow}$ until the self-consistency is attained in the sense that $n_{A\uparrow}=n'_{A\uparrow}$.

It is shown from the calculation that $n_{A\uparrow}$, which meets the self-consistency requirement, is determined uniquely and thus $n_{A\uparrow}=n_{A\downarrow}$. This guarantees $G_{A\uparrow}(E)=G_{A\downarrow}(E)$ and the density of states for an up spin has the same structure as that for a down spin.

In Fig. 5, the calculated density of states is shown. The energy is measured by choosing as an energy unit the Coulomb interaction on site A, i.e., $U_A=U$. The difference between two atomic levels E_B and E_A is taken to be $3U$. The concentration $x=x_A$ of A atoms is chosen to be 0.4. A self-consistent calculation is performed for various values of Δ/U.

As in the case of the Hubbard band for a crystal, we can see that the density-of-states gap comes continuously to zero as Δ/U approaches the critical ratio $(\Delta/U)_c$ from below and that the mobility gap persists even after the ratio Δ/U exceeds the critical ratio $(\Delta/U)_c$ at which the density-of-states gap vanishes. Therefore the actual transition from an insulator to a conductor takes place at a larger ra-

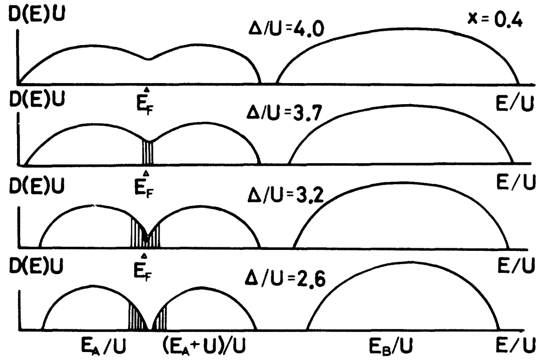

FIG. 5. Calculated density of states $D(E)$ for a disordered binary system in which $U_A=U$, $U_B=0$, $E_B-E_A=3U$, and $x=x_A=0.4$. Results are shown for various values of Δ/U, i.e., $\Delta/U=4.0$, 3.7, 3.2, and 2.6 from the top to the bottom of the figure. The density of states is normalized by $1/U$.

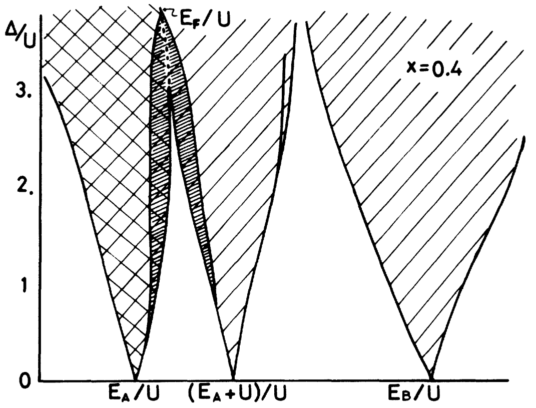

FIG. 6. Dependence of the quasiparticle density of states upon the Δ/U ratio. The concentration of A atoms is $x=x_A=0.4$. The shadowed regions corresponds to localized states. The density of states is nonvanishing in the hatched region of the $(E/U,\ \Delta/U)$ plane. The states in the double-hatched region are occupied.

tio $(\Delta/U)_0$ which corresponds to a disappearance of the mobility gap.

In Fig. 6, the region in which the density of states is nonvanishing is shown in the $(\Delta/U, E/U)$ plane. This is the result for $x=x_A=0.4$. A similar behavior of the density of states as in a regular lattice case studied by Hubbard is observed. The difference is the characteristic property of the mobility edges at the upper band edges in the A bands. Another marked aspect is that, although in the results of Hubbard the two bands are symmetric with respect to energy $E_A+\frac{1}{2}U$ our results show a rather asymmetric band structure. This is reasonable because the existence of the upper B band works to lower these two A bands.

Let us now see how the critical interatomic distances are influenced by the existence of random potentials. For this purpose, we first calculate the lattice constants d_c and d_0 which respectively correspond to $\delta_c=(\Delta/U)_c$ (the ratio at which the density-of-states gap goes to zero) and $\delta_0=(\Delta/U)_0$ (the ratio at which the mobility gap vanishes) on the basis of the nearest-neighbor tight-binding approximation by assuming a simple cubic lattice. Employing a hydrogenlike wave function and remembering that the binding energy for a ground state is almost equal to U, we can relate the bandwidth Δ to the hopping energy in the form

$$\tfrac{1}{2}\Delta=6\times 2\,U(1+d)e^{-d}\ , \tag{4.7}$$

where d is the lattice constant measured in atomic units. Thus, by making use of this equation, we can calculate d_c and d_0 respectively, from $\delta_c=(\Delta/U)_c$ and $\delta_0=(\Delta/U)_0$.

Next, let us define the "corresponding regular lattice" on the idea that the lattice constant $\bar{d}$ of the corresponding regular lattice is equal to the average interatomic distance between metallic atoms in the disordered system under consideration. In the present example, the mean distance between A atoms is taken to be a lattice constant of the corresponding regular system. This yields the following relation for d and $\bar{d}$:

$$Nd^3=N_i\bar{d}^3\ , \tag{4.8}$$

where N is the number of all the lattice points in the original system and N_i the number of the sites occupied by metallic atoms therein. As it is apparent that $N_i=Nx$, we have

$$\bar{d}=d/x^{1/3}\ . \tag{4.9}$$

Thus, we finally can attain the critical ratios $\bar{d}_c$ and $\bar{d}_0$ which are the lattice constants of the respective corresponding regular lattices.

According to the above-described procedure, the critical ratios, the critical interatomic distances, and the lattice constants of the corresponding lattice are calculated and shown in Table I for $x=0.4$. It has been shown in Sec. II that for a completely regular system the critical ratios $\delta_c=(\Delta/U)_c$ and $\delta_0=(\Delta/U)_0$ are, respectively, 2 and $(2+2\sqrt{2})^{1/2}\simeq 2.197$. Assuming the relation (4.5), we derive the lattice constants for these critical ratios such that $d_c\simeq 4.12$ and $d_0\simeq 4.00$.

On comparison of these values with the numerical results in Table I, it is easily concluded that both the density-of-states gap and the mobility gap of a disordered binary alloy disappear for larger interatomic distances than critical lattice constants of a regular lattice.

These results are explained in the following way. Let us first consider the case of critical distance for the density-of-states gap. Suppose we take an arbitrary distance d. If a completely regular lattice has a lattice constant equal to d, then the bandwidth is determined by this value d. On the other hand, if a system is disordered and the average interatomic distance between metallic elements is equal to d, then the bandwidth for this system can be broadened as much as defined by $dx^{1/3}$ and subsequently become wider than that defined by d. So it is possible for some given value of d that the gap

TABLE I. Critical interatomic distances and the lattice constants of the corresponding regular lattice calculated from the critical ratios on the basis of Eq. (4.1). The concentration x of A atoms is chosen to be 0.4.

Critical ratio	Critical interatomic distance	Lattice constant
$\delta_c\cong 3.09$	$d_c\cong 3.56$	$\bar{d}_c\cong 4.89$
$\delta_0\cong 3.82$	$d_0\cong 3.26$	$\bar{d}_0\cong 4.42$

between two density of states of a disordered system has vanished while that for an ordered lattice is still present. The reason why the critical distance for the mobility gap is larger than $d_0 = 4.00$ is easier to understand from physical considerations because the random configuration of metallic atoms works to favor the formation of infinite percolation paths for a given density. The problem in this context will be again discussed in the Sec. V.

V. METAL-NONMETAL TRANSITION IN AMORPHOUS METALS AND LIQUID METALS

Although it is necessary to develop different methods to treat the effect of electron correlations in structurally disordered systems,[23] the present theory which has originally been intended to treat the problem of substitutionally disordered systems may also be applied to structurally disordered systems in the first approximation.[21] For illustration, let us consider the case of $n = 2$ as presented in Sec. IV. When E_B is brought to infinity in the formulation, the model simulates a system where electrons cannot enter sites B because the atomic levels on them are infinite. Therefore, the electronic conduction is possible only through the paths formed by connecting A atoms alone.

As actual systems to which the above idea is applicable, metal–rare-gas mixtures such as Na-Ar[5] and Cu-Ar[6] solids, or liquid metals and alloys in the supercritical region[8,9] are considered. In these materials, the M-NM transitions are experimentally observed and it is suggested that the transitions therein are due to electron correlation as well as the quantum-mechanical percolation mechanism.

We study the same model as described by Eqs. (4.4a) to (4.6) and bring E_B to infinity. We assume a nonmagnetic case with one electron per A atom. As there are only A atoms in the system, the relation $n_{A\uparrow} = n_{A\downarrow} = \frac{1}{2}$ holds for any value of Δ. For convenience of formulation, let us choose, this time, the origin of energy such that $E_A + \frac{1}{2}U = 0$. On making use of the same dimensionless notations as in Sec. IV, the self-consistent equation is written

$$\epsilon = \xi' + \frac{\delta^2 x}{16}\left(\frac{\frac{1}{2}}{\xi' - \frac{1}{2}} + \frac{\frac{1}{2}}{\xi' + \frac{1}{2}}\right) \equiv f(\xi', x) \; . \qquad (5.1)$$

which gives a definition of a function $f(\xi', x)$. The function $f(\xi', x)$ is shown in Fig. 7 as a function of ξ'. The left-hand side of Eq. (5.1) is expressed by a horizontal line at ϵ. As is explained in detail in Ref. 16, the density of states for a given set of parameters ϵ and x is either zero or finite according to whether Eq. (5.1) as a function of ξ' has (a) three real solutions or (b) one real solution in addition to two complex conjugate solutions, respectively. The number of real solutions of Eq. (5.1) is determined by the number of intersections of a horizontal line ϵ with the solid lines corresponding to $f(\xi', x)$. We are interested in the energy region such that $-\frac{1}{2} < \epsilon < \frac{1}{2}$, since the density-of-states gap due to electron correlation, if there exists one at all, comes in this region. The density-of-states gap is persistent if the curve between $\xi' = -\frac{1}{2}$ and $\xi' = \frac{1}{2}$ has a minimum and maximum as indicated in Fig. 7 while it is filled if the curve decreases monotonically. These two cases are characterized by the following statement. Concerning the former case, the curve $f(\xi', x)$ has a maximum and minimum if $df(\xi')/d\xi' > 0$ at the value of ξ' $(-\frac{1}{2} < \xi' < \frac{1}{2})$ which satisfies $d^2 f(\xi')/d\xi'^2 \mid_{\xi' = \xi_0} = 0$. On the other hand, the curve has no extrema if $df(\xi')/d\xi' < 0$ at $\xi' = \xi_0$. Therefore, $df(\xi')/d\xi' \mid_{\xi' = \xi_0} = 0$ gives the critical ratio at which the density-of-states gap disappears.

Observing that

$$\frac{df(\xi')}{d\xi'} = 1 - \frac{\delta^2 x}{16}\left(\frac{\frac{1}{2}}{(\xi' - \frac{1}{2})^2} + \frac{\frac{1}{2}}{(\xi' + \frac{1}{2})^2}\right) \; , \qquad (5.2a)$$

we define a new function $h(\xi', x)$ as

$$h(\xi', x) \equiv \left(\frac{\frac{1}{2}x}{(\xi' - \frac{1}{2})^2} + \frac{\frac{1}{2}x}{(\xi' + \frac{1}{2})^2}\right) \; . \qquad (5.2b)$$

Equation (5.2b) is also shown in Fig. 7 as a function of ξ'. On noting that $f(\xi', x)$ is an odd function of ξ' and $h(\xi', x)$ an even function of ξ', it is easy to see that the minimum of $h(\xi', x)$ exists at $\xi' = \xi_0 = 0$, which enables us to calculate the critical ratio $\delta_c = (\Delta/U)_c$ by using $df(\xi')/d\xi' \mid_{\xi' = 0} = 0$ as

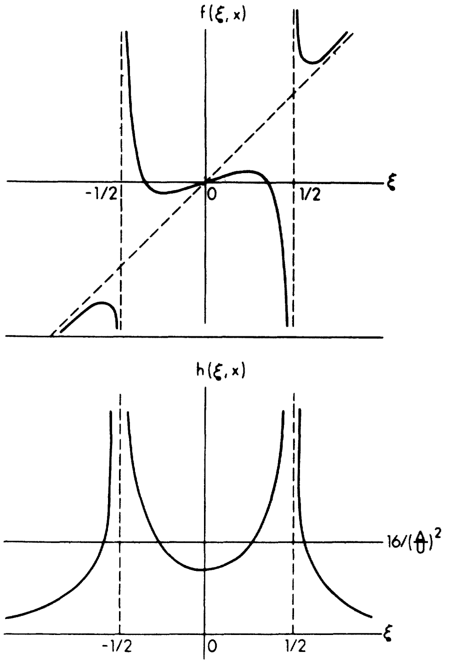

FIG. 7. Functions $f(\xi', x)$ vs ξ' and $h(\xi', x)$ vs ξ'.

$$\delta_c = \left(\frac{16}{h(0,x)}\right)^{1/2} = \frac{2}{x^{1/2}} . \tag{5.3}$$

By definition, this is the ratio at which the density-of-states gap vanishes.

On the other hand, the ratio $\delta_0 = (\Delta/U)_0$ at which the mobility gap becomes zero is evaluated by solving Eq. (5.1) with $\epsilon = 0$ and substituting the obtained solution ξ' into the criterion for localization, i.e., $F(\epsilon) < 1$, where $F(\epsilon)$ is defined by Eq. (2.6). The solution of Eq. (5.1) for $\epsilon = 0$ is written

$$\xi' = i\tfrac{1}{4}(x\delta^2 - 4)^{1/2} \equiv i\beta_0 . \tag{5.4}$$

Remember that we are interested in such a region of δ that $\delta \geq \delta_0 = 2/x^{1/2}$. In our model under consideration, the self-energy Σ is determined by Eq. (4.3). Simple and straightforward algebra yields

$$\begin{aligned}\zeta \equiv \frac{\Sigma}{U} = \xi' + \frac{\delta^2}{16\xi'} &= i\beta_0\left(1 - \frac{\delta^2}{16\beta_0^2}\right) \\ &= -i\,\frac{1}{16\beta_0}\,[4 + (1-x)\delta^2] .\end{aligned} \tag{5.5}$$

By inserting Eq. (5.5) into the criterion $F(\epsilon = 0) < 1$, we obtain an inequality which δ should satisfy, namely,

$$(1 - 6x + x^2)\delta^4 + 8(3 - x)\delta^2 + 16 > 0 . \tag{5.6}$$

It is suggested by this inequality that we have to discuss two regions of x individually. First, let us consider the case where $3 - 2\sqrt{2} < x < 1$. For this case, the required region of δ is expressed as

$$\frac{2}{x^{1/2}} = \delta_c \leq \delta \leq \delta_0 = \frac{2}{[x - (3 - 2\sqrt{2})]^{1/2}} , \tag{5.7}$$

where the density of states at $\epsilon_F = \epsilon = 0$ is nonzero but the states are still localized. For $0 < x < 3 - 2\sqrt{2}$, the states at the Fermi level are always localized no matter how large $\delta = \Delta/U$ may be.

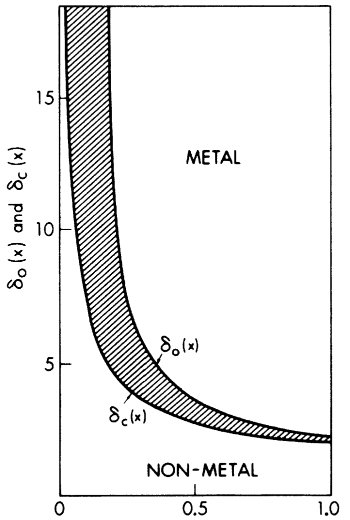

FIG. 8. Dependence of the two critical ratios $(\Delta/U)_c$ and $(\Delta/U)_0$ upon the concentration x of metallic elements. The shadowed region indicates that the density of states at the Fermi level is nonvanishing for these values of x and Δ/U but the states are localized.

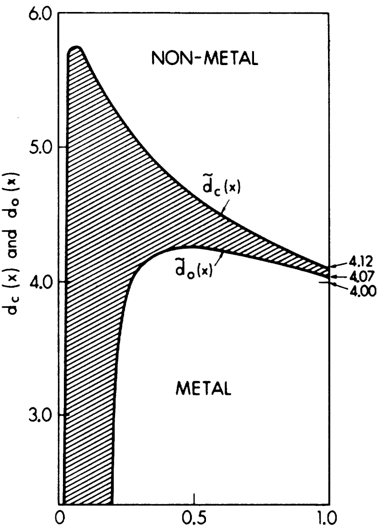

FIG. 9. Dependence of the two critical interatomic distances $\tilde{d}_c$ and $\tilde{d}_0$ upon the concentration x of metallic elements. Detailed definitions of $\tilde{d}_c$ and $\tilde{d}_0$ are given in the text.

The critical values δ_c and δ_0 for the disappearances of the density-of-states gap and the mobility gap, respectively, are shown in Fig. 8. The shadowed region corresponds to the states of the Fermi level ($\epsilon_F = \epsilon = 0$) such that the density of states is nonzero but the states are localized. Note that the system is nonmetallic in this region of (x, δ). In the upper side of the (x, δ) plane, the density of states is nonzero at ϵ_F and the states thereof are extended, accordingly the system being metallic; while in the lower side of the (x, δ) plane, the Fermi level is in the density-of-states gap and therefore the system is an insulator.

It is interesting to note that the value $x_c = 3 - 2\sqrt{2}$ is nearly the same as the percolation concentrations for several typical three-dimensional lattices. The result that the states at the Fermi level are all localized for x smaller than x_c may be understood to indicate that no infinite percolation path can be formed for $x < x_c$.

Another important conclusion is drawn by evaluating the lattice constants $\tilde{d}_c$ and $\tilde{d}_0$ of the corresponding regular lattice based upon the concept and formulations developed in Sec. IV. The results of a numerical calculation are plotted in Fig. 9 as functions of x. Again, the shadowed region denotes that the states there are localized although the density of states is not zero. Note that the critical lattice constants should be all the same as the values at $x = 1$ if these corresponding lattices *are* regular.

Therefore it is suggested from the figure that both critical values are larger than the completely regular lattice except for those regions where $x < x_c$. The explanation for these results has been given at the end of Sec. IV.

VI. DISCUSSION

It is generally accepted that the M-NM transition in a regular lattice with a half-filled s band is induced by the change of the lattice constant and that the mechanism of the transition is the effect of the Coulomb interactions between electrons. It is pointed out in this paper that even in regular systems potential fluctuations due to the random configurations of spins do exist and it is essential to take these potential fluctuations into account in order to discuss qualitative behaviors of the transition.

The M-NM transitions in substitutionally disordered systems with half-filled s bands are shown to be mainly due to electron correlation but the effect of potential fluctuations is also important. Especially when the potential fluctuations are very large, the localization of electrons in the Anderson sense plays an essential role. The potential fluctuations in disordered systems are due to random configurations of spins as well as random distributions of atoms, and measured by, for instance, in the model studied in Secs. IV and V, U, the Coulomb interaction between opposite spins on the same site, $v = |E_A - E_B|/U$, a normalized energy difference between the A and B atomic levels, and x, the concentration of A atoms. Larger values of U, v, and x^{-1} produce a larger fluctuation and consequently a smaller interatomic distance is required so that the system may attain metallic properties. An important point is that, although the behavior of the electron conductivity or mobility in the metallic phase might be explained, for instance, by using the percolation theory, the formation of an infinite percolation path is possible only when the density of metallic atoms is high enough to make the density-of-states gap in the Mott-Hubbard band disappear. In other words, metallic conduction is attained only when both conditions for the so-called Mott transition and the percolation conduction are fulfilled, and it is expected from our results that the requirement for the Mott transition is first satisfied accompanying the increase of the density or equivalently the decrease of the average interatomic distance, and a higher density or a small interatomic distance is required to meet the condition for the formation of an infinite percolation path.

Although the present theory is originally intended to treat electron correlations in substitutionally disordered systems, it also serves as an approximate theory to discuss the problem of the M-NM transitions in topologically or structurally disordered systems. This alloy analogy of an amorphous or liquid metal corresponds to the case with $v = \infty$ and x small in the binary-alloy model described in the above, and extracts some essential features of the problem. Namely, it has been shown that, no matter how small the interatomic distance is, electrons in these systems are always localized when the concentration x of metallic elements is smaller than some critical value. This indicates that the infinite potential $v = \infty$ prohibits electrons from occupying B atom sites and therefore percolation paths cannot be connected infinitely when x is small enough. It must be noted however that the positions of atoms are restricted to regular lattice sites in the alloy analogy model of structurally disordered systems, and this restriction might not be appropriate in actual amorphous or liquid metals because structural randomness in the atomic distribution of these systems is usually expected to favor the formation of infinite percolation paths. Therefore, in structurally disordered systems, the mobility gap may disappear at a smaller value of δ than estimated by the present theory and accordingly the system attains the transition from an insulator to a conductor at a larger interatomic distance. This subject will be discussed elsewhere at length.[23]

ACKNOWLEDGMENTS

The authors are grateful to Professor H. Endo for fruitful discussions. One of the authors (F.Y.) is also indebted to Dr. D. Mattis for useful advice and warm encouragement.

---

[*] The main part of this paper was reported at the "Kyoto Seminar" held in Kyoto, 30 August–1 September 1972. The proceedings have been published.

[†] Research at Yeshiva supported in part under Grant Nos. AFOSR-73-2430 and AFOSR-72-2153B.

[‡] On leave from Department of Applied Physics, Tokyo Institute of Technology, Meguroku, Tokyo 152, Japan.

[1] P. W. Anderson, Phys. Rev. **109**, 1492 (1958).

[2] N. F. Mott, Philos. Mag. **24**, 935 (1971). N. F. Mott and E. A. Davis, *Electronic Process in Noncrystalline Materials* (Clarendon, Oxford, England, 1972). References of a series of works by N. F. Mott will be found therein.

[3] F. Yonezawa and M. Watabe, International Seminar on Electron in Disordered Systems (1973) (unpublished), p. 72.

[4] See, for instance, *Solutions Metal-Ammoniac*, edited by G. Leputre and M. J. Sienko (Benjamin, New York, 1964).

[5] R. C. Cate, J. G. Wright, and N. E. Cusack, Phys. Lett. **32A**, 469 (1970).

[6] H. Endo, J. G. Wright, A. Eatah, and N. E. Cusack, J. Phys. Soc. Jap. **34**, 666 (1973).

[7] B. Raz, A. Gedanken, U. Even, and J. Jortner, Phys. Rev. Lett. **28**, 1643 (1972).

[8] See, for instance, R. G. Ross and D. A. Greenwood, Prog. Mater. Sci. **14**, 173 (1969).

[9] For more recent developments, see the *Proceedings of the Second International Conference on the Properties of Liquid Metals* (Taylor & Francis, New York, 1973).

[10] N. F. Mott, Proc. Phys. Soc. Lond. **62**, 416 (1949).

[11] J. Hubbard, Proc. R. Soc. Lond. **281**, 401 (1964).

[12] For instance, see a review article by F. Yonezawa and K. Morigaki, Progr. Theor. Phys. (Kyoto) (to be published).

[13] N. F. Mott, Adv. Phys. **16**, 49 (1967).

[14] M. H. Cohen, H. Fritsche, and S. R. Ovshinsky, Phys. Rev. Lett. **22**, 1065 (1969).

[15] F. Yonezawa and M. Watabe, Solid State Commun. **11**, 1667 (1972).

[16] F. Yonezawa, Phys. Rev. B **7**, 5170 (1973).

[17] N. F. Mott, Philos. Mag. **24**, 935 (1971).

[18] M. Cyrot, Philos. Mag. **25**, 1031 (1972).

[19] E. N. Economou and M. H. Cohen, Phys. Rev. B **5**, 2931 (1972).

[20] N. F. Mott and E. A. Davis, Philos. Mag. **17**, 1269 (1968).

[21] H. Fukuyama and H. Ehrenreich, Phys. Rev. B **7**, 3266 (1973).

[22] J. M. Ziman, J. Phys. C **2**, 1230 (1969).

[23] F. Yonezawa, M. Watabe, M. Nakamura, Y. Ishida, and T. Ogawa, in Ref. 9, p. 373.