Coherent-Potential-Approximation Calculation on the Falicov-Kimball Model of the Metal-Insulator Transition*

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We solve the Falicov-Kimball model of the metal-insulator transition in the coherent-potential approximation. This yields exact results in certain limits in which molecular field theory is incorrect, and it is a better approximation over the whole range of the parameters. We find that, at least below the point at which bound states appear, all phase transitions vanish.

In a recent paper Falicov and Kimball[1] have proposed a simple model capable of accounting for a wide range of transitions found experimentally in transition-metal and rare-earth oxides. In the simplest case[2] the model, when treated in the molecular-field-theory (MFT) or virtual-crystal approximation, exhibits, when the strength of the electron-hole interaction is varied, the following range of behavior: (a) an insulating phase with no transitions, (b) a continuous metal-to-insulator transition, (c) a first-order metal-to-insulator transition, and (d) a metallic phase at all temperatures. The model has since been extended[3,4] to include magnetic interactions and has been found qualitatively successful in describing the phases found in the system $(V_{1-x}Cr_x)_2O_3$. In this paper we report the results of a coherent-potential-approximation (CPA) calculation on the nonmagnetic model. The CPA of Soven[5] has been found successful in a wide range of alloy problems and has also been previously used by Hubbard[6] in an electron correlation problem. In this case the CPA yields exact results in several limiting cases where MFT gives incorrect results, and it is a better approximation over the whole range of the parameters.

The model consists of localized atomic levels and a conduction band separated by a gap Δ. From any atom only one electron may be removed, and the conduction electrons interact with the holes via an attractive interaction of strength G localized within one atomic cell. We consider, therefore, the following Hamiltonian[2]:

$$\mathcal{H} = \sum_{k,\sigma} [\epsilon(k) + \Delta + \tfrac{1}{2}W] C_{k\sigma}{}^\dagger C_{k\sigma} + E \sum_{i,\sigma} b_{i\sigma}{}^\dagger b_{i\sigma} - G \sum_{i\sigma\sigma'} b_{i\sigma}{}^\dagger C_{i\sigma'}{}^\dagger C_{i\sigma'} b_{i\sigma}, \qquad (1)$$

where $C^\dagger$ and C are conduction-electron creation and annihilation operators and $b^\dagger$ and b are hole creation and annihilation operators. The energy E, which we take to be zero, is the energy needed to remove an electron from an atom; W is the bandwidth, $W = \max\epsilon(k) - \min\epsilon(k)$, which we fix at unity; Δ is the gap between the valence and conduction bands; G is the strength of the electron-hole interaction which we always take to be greater than 0. In MFT one replaces the operator $\sum_\sigma b_{i\sigma}{}^\dagger b_{i\sigma}$ by its mean value $\sum_\sigma \langle b_{i\sigma}{}^\dagger b_{i\sigma}\rangle = n_T$, where n_T is the fraction of ionized atoms at temperatures T. The Hamiltonian then becomes

$$\mathcal{H} = \sum_{k,\sigma} [\epsilon(k) + \Delta + \tfrac{1}{2}W - Gn_T] C_{k\sigma}{}^\dagger C_{k\sigma}. \qquad (2)$$

To obtain the thermodynamics we calculate the free energy per atom $F = U - TS$:

$$U = 2 \int_{-W/2}^{W/2} d\omega\, \rho_0(\omega)(\omega + \Delta + \tfrac{1}{2}W - Gn_T)n(\omega), \qquad (3)$$

$$S = -2k \int_{-W/2}^{W/2} d\omega\, \rho_0(\omega)\{n(\omega)\ln n(\omega) + [1 - n(\omega)]\ln[1 - n(\omega)]\} - k[n_T \ln n_T + (1 - n_T)\ln(1 - n_T) - n_T \ln q], \qquad (4)$$

where $n(\omega)$ is a function to be determined by setting $\partial F/\partial n(\omega) = 0$. The first term of (4) is the conduction-band entropy, the second term the atomic entropy. The factor of $q \equiv (2J + 1)/(2J_0 + 1)$, the ratio of an ionized atom's spin multiplicity to the spin multiplicity of an un-ionized atom, is henceforth taken to be 2. The function $n(\omega)$ must satisfy the condition

$$n_T = 2 \int_{-\infty}^{\infty} d\omega\, \rho_0(\omega)n(\omega), \qquad (5)$$

where the factor 2 comes from summation over conduction-electron spin states. $\rho_0(\omega)$ is the unper-

turbed density of states, which obeys the condition $\int_{-\infty}^{\infty} d\omega\, \rho_0(\omega) = 1$. In all calculations we take a simple cubic density-of states which we approximate, for $W = 1$, by

$$\rho_0(\omega) = \begin{cases} \frac{3}{2}[1 - \pi^{-1}\cos^{-1}(6\omega + 2)], & -\frac{1}{2} \leqslant \omega < -\frac{1}{6}, \\ \frac{3}{2}, & -\frac{1}{6} \leqslant \omega < \frac{1}{6}, \\ \frac{3}{2}\pi^{-1}\cos^{-1}(6\omega - 2), & \frac{1}{6} \leqslant \omega < \frac{1}{2}, \\ 0, & |\omega| \geqslant \frac{1}{2}. \end{cases} \tag{6}$$

Equation (5) simply expresses the fact that the number of holes is equal to the number of conduction electrons. Minimizing F with respect to $n(\omega)$ and using (5) we obtain

$$n(\omega) = \left\{ \frac{n_T}{q(1 - n_T)} \exp\left[\beta\left(\omega + \Delta + \frac{W}{2} - 2Gn_T\right)\right] + 1 \right\}^{-1}. \tag{7}$$

Substituting (7) into (5) we obtain an equation for n_T as a function of temperature. For the simple cubic density of states the value of G at which electron-hole bound states begin to form is[2] $G = 0.329\,73W$. We must restrict G to less than this value if we want to interpret the excited electrons as carriers. The MFT equations yield the curves, taken from Ref. 2, of n_T as a function of the reciprocal temperature β shown in Fig. 1. In each case $\Delta = \frac{1}{70}$ and G is varied. Curve A shows an insulating phase, curve B a first-order metal-insulator transition, and curve C a metallic phase at all temperatures. The transition and the metallic phase are made possible by the existence of more than one solution of Eqs. (5) and (7) at the same temperature. The physical solution is the one with the lower free energy.

In the CPA calculation we also replace the operator $\sum_\sigma b_{i\sigma}^{\dagger} b_{i\sigma}$ by a c-number which we allow to be a random variable restricted to the values 0 and 1. The Hamiltonian then is

$$\mathcal{H} = \sum_{k,\sigma} [\epsilon(k) + \Delta + \tfrac{1}{2}W]\, C_{k\sigma}^{\dagger} C_{k\sigma} - \sum_{i,\sigma} G_i\, C_{i\sigma}^{\dagger} C_{i\sigma}, \tag{8}$$

where $G_i = G$ with probability n_T and $G_i = 0$ with probability $1 - n_T$. The Hamiltonian describes an electron moving in a lattice of random potentials and we apply the CPA[5] to it. The one-particle configuration-averaged Green's function has the form

$$\mathcal{G}(k, \omega) = [\omega - \epsilon(k) - \Sigma(\omega)]^{-1}, \tag{9}$$

where $\Sigma(\omega)$ is determined by the equation

$$\Sigma + n_T G + \Sigma(\Sigma + G)F(\omega) = 0, \tag{10}$$

with

$$F(\omega) = \int_{-\infty}^{\infty} d\omega'\, \rho_0(\omega')[\omega - \Sigma(\omega) - \omega']^{-1}. \tag{11}$$

The effect of the electron-hole attraction is then entirely included in the modified density of states which is given by

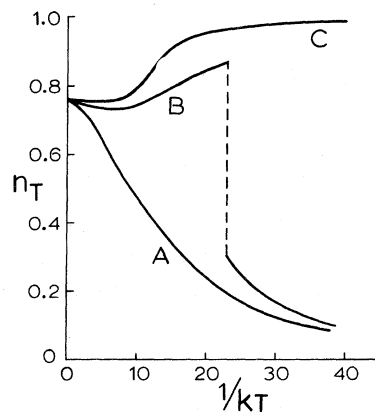$$\rho(\omega) = -\pi^{-1}\mathrm{Im}F(\omega). \tag{12}$$

FIG. 1. Plot of the MFT curves of n_T as a function of $1/kT$ for $W = 1$ and $\Delta = \frac{1}{70}$. Curve A, $G = 0.25$; curve B, $G = 21.4/70$; curve C, $G = \frac{23}{70}$. These curves are taken from Ref. 2.

To facilitate the numerical solution of Eq. (10) we have linearly interpolated the unperturbed density of states in the calculation of the function $F(\omega)$ as suggested by Kirkpatrick, Velický, and Ehrenreich.[7]

The CPA is a better approximation than MFT over the whole range of parameters. In particular it is exact in the so-called atomic limit, $G \gg W$. The CPA Green's function in this case is

$$g(\omega) = \frac{n_T}{\omega - \epsilon + G} + \frac{1 - n_T}{\omega - \epsilon},$$ (13)

where ϵ is the energy of the infinitely narrow band. In MFT only the single pole at $\omega = \epsilon - Gn_T$ is obtained.

To obtain the thermodynamics we introduce the chemical potential μ which is determined by the condition

$$n_T = 2 \int d\omega \, \rho(\omega)\{1 + \exp[\beta(\omega + \Delta + \tfrac{1}{2}W - \mu)]\}^{-1}.$$ (14)

The total free energy is then

$$F(n_T, T) = -2kT \int d\omega \, \rho(\omega) \ln\{1 + \exp[-\beta(\omega + \Delta + \tfrac{1}{2}W - \mu)]\}$$
$$+ n_T \mu + kT [n_T \ln(n_T/q) + (1 - n_T) \ln(1 - n_T)].$$ (15)

The equilibrium state at temperature T is then obtained by minimizing F with respect to n_T. The results are shown in Fig. 2 for the same values of G as those producing the curves of Fig. 1 in MFT. In all cases n_T is a smooth function of β. The curves A and B have the limit $n_T \to 0$ as $T \to 0$ whereas curve C has the limit $n_T \approx 0.55$ as $T \to 0$. Both metallic and insulating phases are therefore found but no phase transitions occur. The first-order phase transitions found in MFT result from the existence of two minima in the function $F(n_T, T)$ which cross at some temperature. With the CPA we find, at all temperatures, only one minimum in F as n_T is varied from 0 to 1.

The results of the MFT version of the Falicov-Kimball model are qualitatively successful in accounting for a large variety of experimental data. It i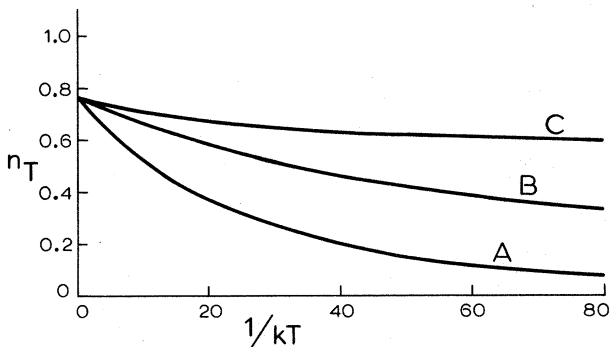s tempting, therefore, to search for ways of modifying the Hamiltonian[1] to make MFT a better approximation. An obvious possibility is to increase the range of the electron-hole interaction beyond a single cell. The CPA used here is valid only for point scattering and it is clear that for a longer-range interaction, MFT will become the preferred approximation. This would have the added feature of reducing the size of the potential G needed to bring about phase transitions and would allow for larger gaps between the valence and conduction band.

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FIG. 2. Plot of the CPA curves of n_T as a function of $1/kT$ for $W = 1$, $\Delta = \frac{1}{70}$. Curve A, $G = 0.25$; curve B, $G = 21.4/70$; curve C, $G = \frac{23}{70}$.

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