In the free-electron model one finds agreement with the result of $1/\tau_m$ deduced from experiment [Eq. (10)] if $|J|=0.1$ eV. This value of J is in reasonable agreement with results of a free-electron model interpretation of the superconducting transition temperature suppression ($|J|=0.073$ eV) found by Maple[14] in magnetically dilute $La_{1-c}Gd_cAl_2$.

We believe that this is the first observation of the asymptotic $t^{1/2}$ limit of diffusionless relaxation in a metal. The simplifying factors we have discussed enable us to identify unambiguously the impurity-process relaxation as arising from longitudinal dipolar fluctuations. Although the functional dependence is complicated in previous studies[2-4] by the possible temperature variation of r_0, we find that the magnitude and functional dependence of the high-field ($\omega_m\tau_m>1$) data in each of these cases can be accounted for by the same mechanism.[15]

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[1] This topic has been reviewed in detail by I. J. Lowe and D. Tse, Phys. Rev. 166, 279 (1968). See also J. I. Kaplan, Phys. Rev. B 3, 604 (1971).

[2] R. E. Levine, Phys. Lett. 28A, 504 (1969).

[3] P. Bernier, H. Launois, and H. Alloul, J. Phys. (Paris) 32, C1-513 (1971).

[4] F. Fradin, Phys. Rev. Lett. 26, 1033 (1971).

[5] A review of these processes is given in B. Giovannini, P. Pincus, G. Gladstone, and A. J. Heeger, J. Phys. (Paris) 32, C1-163 (1971).

[6] This expression is directly obtained from an analysis similar to that of D. Tse and S. R. Hartmann, Phys. Rev. Lett. 21, 511 (1968). The details of its derivation and an extension to arbitrary concentration will be presented elsewhere.

[7] K. Yosida, Phys. Rev. 106, 893 (1957).

[8] The solution to the diffusion equation in the rapid-diffusion limit (Ref. 1) depends on a diffusion barrier that also has the same difficulties as r_0.

[9] A. Redfield and W. Yu, Phys. Rev. 169, 443 (1968).

[10] J. Korringa, Physica (Utrecht) 16, 601 (1950).

[11] A. Narath, Phys. Rev. 162, 320 (1967).

[12] This partition of the magnetization recovery is not immediately obvious since, as indicated in Ref. 5, interference between two relaxation processes can occur in certain cases. In this system, however, the sources of the hyperfine fields (a contact interaction for the conduction electrons and a dipolar field for the impurity) are distinct and the processes are independent.

[13] H. E. Rorschach, Physica (Utrecht) 30, 38 (1964).

[14] M. B. Maple, Solid State Commun. 8, 1915 (1970).

[15] Comments on Ref. 4 are to be published elsewhere.

Self-Consistent Theory of Clusters in Disordered Alloys

F. Cyrot-Lackmann
Institut Laue-Langevin, 38-Grenoble, France

and

F. Ducastelle
ONERA, 29 avenue de la Division Leclerc, 92-Chatillon, France

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A way to deal with cluster effects in disordered alloys is given through an extension of the coherent-potential–approximation theory. An explicit expression is derived for pair effects and is shown to give the exact c^2 term in the lower concentration limit.

The coherent-potential approximation (CPA) is known to be what might be called the best single-site approximation in the disordered-alloy problem. Our purpose here is to generate higher approximations which give self-consistent solutions for pairs, triplets, etc. Some attempts along these lines have recently been made through various techniques[1-4]; as we shall notice later on, they have failed to give the correct behavior in the low-concentration limit. This point will be fully discussed elsewhere.

Here we give an extension of the formalism developed by Velický, Kirkpatrick, and Ehrenreich[5] in the single-site approximation to take into account pairs of atoms or larger clusters. We start from the usual Hamiltonian H describing an alloy $A_{1-c}B_c$:

$$H=H_0+V, \tag{1}$$

with

$$H_0=\sum_{n\neq m}|n\rangle\beta_{nm}\langle m|, \tag{2}$$

$$V=\sum_n|n\rangle\epsilon_n\langle n|, \tag{3}$$

where ϵ_n is the random atomic level, the value of which is ϵ_A or ϵ_B; the overlap integral β_{nm} is assumed to be translationally invariant.

The Green's function G is

$$G=(Z-H)^{-1}=(Z-H_0-V)^{-1}. \tag{4}$$

The problem is to calculate the average of G over all the configurations of the alloy,

$$\langle G\rangle=\langle(Z-H)^{-1}\rangle. \tag{5}$$

The self-energy Σ is then defined by

$$\langle G\rangle=(Z-H_0-\Sigma)^{-1}. \tag{6}$$

Using a notation similar to that of Velický, Kirkpatrick, and Ehrenreich,[5] G can be related to $\langle G\rangle$ through the T matrix:

$$G=\langle G\rangle+\langle G\rangle T\langle G\rangle, \tag{7}$$

where

$$T=(V-\Sigma)[1-\langle G\rangle(V-\Sigma)]^{-1}. \tag{8}$$

Hence, we obtain the self-consistent relationship for Σ,

$$\langle T\rangle=0. \tag{9}$$

Let us also define V_n, Σ_n, T_n, and Q_n by

$$V_n=|n\rangle\epsilon_n\langle n|. \tag{10}$$

$$\Sigma_n=|n\rangle\langle n|\Sigma, \tag{11}$$

$$Q_n=|n\rangle\langle n|T. \tag{12}$$

$$T_n=(V_n-\Sigma_n)[1-\langle G\rangle(V_n-\Sigma_n)]^{-1}. \tag{13}$$

We may then write

$$Q_n=T_n[1+\langle G\rangle\sum_{m\neq n}Q_m]. \tag{14}$$

From (9) and (12) we have the exact relation

$$\langle Q_n\rangle=0. \tag{15}$$

The CPA is given by decoupling the average on the right-hand side of (14), that is,

$$\langle Q_n\rangle=\langle T_n\rangle[1+\langle G\rangle\sum_{m\neq n}\langle Q_m\rangle], \tag{16}$$

which gives

$$\langle T_n\rangle=0. \tag{17}$$

The neglecting of fluctuations such as $\langle T_n\langle G\rangle Q_m\rangle-\langle T_n\rangle\langle G\rangle\langle Q_m\rangle$ is obviously equivalent to taking account of only repeated scattering by a single site.[5] To go further, we iterate (14) in such a way that repeated scattering by all pairs of atoms appears explicitly.

Let us write

$$Q_n=T_n+\sum_{m\neq n}R_{nm} \tag{18}$$

with

$$R_{nm}=T_n\langle G\rangle Q_m. \tag{19}$$

We have

$$\begin{aligned}R_{nm}&=T_n\langle G\rangle(T_m+\sum_{l\neq m}T_m\langle G\rangle Q_l)\\&=T_n\langle G\rangle T_m(1+\sum_{l\neq m,n}\langle G\rangle Q_l)+T_n\langle G\rangle R_{mn}.\end{aligned} \tag{20}$$

In the same way

$$R_{mn}=T_m\langle G\rangle T_n(1+\sum_{l\neq m,n}\langle G\rangle Q_l)+T_m\langle G\rangle R_{nm}. \tag{21}$$

The system (20)–(21) is easily solved:

$$R_{nm}=P_{nm}[1+\sum_{l\neq m,n}\langle G\rangle Q_l] \tag{22}$$

with

$$\begin{aligned}P_{nm}=&[1-T_n\langle G\rangle T_m\langle G\rangle]^{-1}\\&\times T_n\langle G\rangle T_m[1+\langle G\rangle T_n].\end{aligned} \tag{23}$$

Decoupling the average in $\langle R_{nm}\rangle$, one then gets a pair approximation, as P_{nm} describes the scattering by the pair n-m:

$$\langle R_{nm}\rangle=\langle P_{nm}\rangle(1+\sum_{l\neq m,n}\langle G\rangle\langle Q_l\rangle), \tag{24}$$

and from (15) and (18), one gets

$$\langle R_{nm}\rangle=\langle P_{nm}\rangle \tag{25}$$

and

$$\langle T_n+\sum_{n\neq m}P_{nm}\rangle=0. \tag{26}$$

Equation (26) represents the self-consistent relationship for the pair approximation, the analog to (17) for the CPA.

Let us now briefly comment on this pair approximation. The self-energy Σ and hence the T_n matrix are no longer diagonal in the site representation as they were in the CPA. This off-diagonality leads to much more difficult numerical computations which are currently being studied.

Next, we discuss the low-concentration limit. In this limit it is rather easy to get the exact form of Σ up to second-order terms in c, the concentration of B impurities. If G^0 is the pure A-metal Green's function, we have

$$\langle G\rangle=G^0+G^0\langle vG\rangle \tag{27}$$

with

$$v = \sum_n |n\rangle(\epsilon_n - \epsilon_A)\langle n|,$$
$$= \sum_n (V_n - V_n^A), \tag{28}$$

and

$$V_n^A = |n\rangle\epsilon_A\langle n|. \tag{29}$$

Defining new operators $\mathcal{T}$ and t_n by

$$\mathcal{T} = v(1 - G^0 v)^{-1}, \tag{30}$$

$$t_n = (V_n - V_n^A)[1 - G^0(V_n - V_n^A)]^{-1}, \tag{31}$$

we can expand $\mathcal{T}$ just like T. The decoupling procedure (24) is exact up to second-order terms in c. Thus

$$\langle\mathcal{T}\rangle = c\sum_n t_n + c^2 \sum_{\substack{n\\ m\neq n}} (1 - t_n G^0 t_m G^0)^{-1} \times t_n G^0 t_m (1 + G^0 t_n) + O(c^3). \tag{32}$$

$\langle\mathcal{T}\rangle$ is related to Σ by

$$\Sigma = \langle\mathcal{T}\rangle(1 + G^0\langle \mathrm{T}\rangle)^{-1}. \tag{33}$$

Thus

$$\langle n|\Sigma|n\rangle = \langle 0|\Sigma|0\rangle$$
$$= ct_0 + c^2\left[-t_0^2 G_{00}^0 + \sum_{n\neq 0} \frac{t_0^3 G_{0n}^0 G_{n0}^0}{1 - t_0^2 G_{0n}^0 G_{n0}^0}\right], \tag{34}$$

$$\langle 0|\Sigma|i\rangle = c^2 \frac{t_0^4 (G_{0i}^0)^2 G_{i0}^0}{1 - t_0^2 G_{0i}^0 G_{i0}^0}, \quad i \neq 0,$$

where

$$G_{ij}^0 = \langle i|G^0|j\rangle \text{ for all } i,j.$$

In the CPA Σ, which is site diagonal, is exact to first order in c, that is,

$$\langle 0|\Sigma|i\rangle = ct_0\delta_{0i} + O(c^2). \tag{35}$$

Further, by expanding the expression (26) as a function of c, it is easily verified that the exact expression (32) is obtained.

If we examine the pair approximations given previously by various authors, it is found that they failed to give the correct behavior at low concentration for various reasons which will be examined elsewhere.[6]

A similar discussion can be given for other limits. For example, it has been verified that pair terms are exact in our approximation for the atomic and virtual crystal limits, $\beta_{mn} \to 0$, and $\epsilon_B - \epsilon_A \to 0$, respectively. It can also be shown that the eleven first moments of the density of states are exact in this pair approximation, while in the CPA case only the seven first moments are exact.[7] There is no basic difficulty in generating approximations of any higher order. For example, for triplets we write

$$Q_n = T_n + \sum_{m\neq n} R_{nm} + \sum_{\substack{m\neq n\\ l\neq m,n}} S_{nml} \tag{36}$$

with

$$S_{nml} = P_{nm}\langle G\rangle Q_l. \tag{37}$$

Thus we obtain a system of six equations for the six S matrices corresponding to the six permutations of n, m, and l. The solution is straightforward but rather tedious, and we shall not write its expression here. A self-consistent relationship for triplets follows in the same way as for pairs.

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[1] R. N. Aiyer, R. J. Elliott, J. A. Krumhansl, and P. L. Leath, Phys. Rev. 181, 1006 (1969).

[2] K. F. Freed and M. H. Cohen, in "Electron Density of States," edited by L. H. Bennett, National Bureau of Standards Special Publication No. 323 (U. S. GPO, Washington, D. C., to be published).

[3] M. Tsukada, J. Phys. Soc. Jap. 26, 684 (1969).

[4] V. Čápek, Phys. Status Solidi 43, 61 (1971).

[5] B. Velický, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. 175, 747 (1968).

[6] P. Soven, Phys. Rev. 178, 1136 (1969), however has given some pair corrections to the CPA which give the correct behavior as $c \to 0$, but not in a self-consistent way.

[7] F. Brouers, private communication.