KFKI-1980-93

B. VASVÁRI

ELECTRON DENSITIES OF LIQUID AND AMORPHOUS METALS

Hungarian Academy of Sciences

CENTRAL
RESEARCH
INSTITUTE FOR
PHYSICS

BUDAPEST

2017

KFKI-1980-93

ELECTRON DENSITIES OF LIQUID AND AMORPHOUS METALS

B. Vasvári

Central Research Institute for Physics
H-1525 Budapest 114, P.O.B. 49, Hungary

To appear in the Proceedings of the Conference on Metallic Glasses: Science and Technology, Budapest, Hungary, June 30 - July 4, 1980; Paper E-18

HU ISSN 0368 5330
ISBN 963 371 739 6

АННОТАЦИЯ

С использованием математических методов, применяемых для расчета структуры полос кристаллических материалов, разработан метод определения локальных плотностей электронных состояний в структурно-неупорядоченных металлах. Целью разработки являлась разработка простого, но реально описывающего алгоритма для определения плотностей состояний. Вследствие изотропности этих систем имеется возможность существенного упрощения, если при расчетах до конца придерживаться представлению данных в форме координат. Приведены предварительные цифровые результаты для аморфного железа.

KIVONAT

Egy eljárást fejlesztettünk ki a szerkezetileg rendezetlen fémek lokális elektron-állapotsürüségeinek (DOS) meghatározására a kristályos anyagok sávszerkezetének számitására szokásos matematikai módszerek felhasználásával. A célkitüzés egy egyszerü, mégis realisztikus algoritmus kifejlesztése az állapotsürüség meghatározására. A szóbanforgó rendszerek izotrópiájának következtében lényeges egyszerüsitési lehetőségek adódnak, ha a számitások során mindvégig a koordináta-reprezentációban maradunk. Előzetes numerikus eredményeket mutatunk be az amorf vasra vonatkozóan.

ABSTRACT

A formalism is developed to calculate the local density of states (DOS) in a structurally disordered metal using the mathematical techniques of band structure calculations of crystalline materials. The aim is to develop a simple, but realistic algorithm for the calculation of the DOS. Remaining consistently in the direct space throughout the whole calculation, a considerable simplification occurs due to the isotropy of the systems involved. Preliminary results for amorphous iron are presented.

INTRODUCTION

Realistic calculations for the electronic density of states (DOS) of structurally disordered metals usually apply the Green's function formalism and the muffin-tin approximation [1]. Similarly to the case of crystalline metals, expansions in the momentum ($\underline{k}$) space and angular momentum ($L = (\ell,m)$) representations lead to the Korringa-Kohn-Rostocker (KKR) type of formulas and an indirect relationship between the quasi-particle energy, E, and the wave number vector, $\underline{k}$, in the reciprocal space. Due to the lack of translational invariance in amorphous metals, the wave number vector, $\underline{k}$, is not a good quantum number for the one electron states, therefore, complications occur, like complex wave number vectors, finite lifetimes in the state $\underline{k}$, etc. In this paper, we do also use the Green's function formalism and the muffin-tin approximation, but without introducing any wave-number dependence into our formalism, in other words, we use consistently the direct, coordinate representations of our quantities. As a consequence we get rather simple results for the density of states in structurally disordered systems like liquid metals or metallic glasses.

A SURVEY OF THE MATHEMATICAL TREATMENT

In the following only a short outline of the mathematical treatment can be given. The starting point is an expression for the local DOS, $g(\underline{r},E)$, in the terms of immaginary part of the ensemble averaged Green's function of the total system:

$$g(\underline{r},E) = -\frac{1}{\pi}\,\mathrm{Im} < G(\underline{r},\underline{r},E)>. \qquad (1)$$

It is supposed that the potential of the amorphous system can be written as superposition of non-overlapping atomic-like potentials:

$$V(\underline{r}) = \sum_{\underline{R}_i} v_i(\underline{r}-\underline{R}_i).$$

Here $v_i(\underline{r}-\underline{R}_i)$ is the muffin-tin potential around the i-th atom centered at the position $\underline{R}_i$.

It is well known, the Green's function can be expanded in the terms of the t-matrices, t_i, of the individual scatterers

$$\begin{aligned} G = G_o &+ \sum_i G_o t_i G_o + \sum_i G_o t_i \cdot \sum_{j\neq i} G_o t_j \cdot G_o \\ &+ \sum_i G_o t_i \cdot \sum_{j\neq i} G_o t_j \cdot \sum_{\ell\neq j} G_o t_\ell \cdot G_o + \ldots \end{aligned} \qquad (2)$$

where G_o is the free-electron propagator. Using a simple change of the variables, the $\underline{R}_i$ dependence can be transformed from $t_i = t(\underline{r}-\underline{R}_i, r'_i-\underline{R}_i)$ to the G_o's: $G_o \longrightarrow G_{ij} \equiv G_o(\underline{r}-\underline{r}'-\underline{R}_i+R_j)$. Next we suppose that we have a simple, one component system, and assume, that the scattering properties of the individual atoms are all the same, i.e., the t_i matrices are independent of which atom is considered. This assumption is equivalent to the average t-matrix approximation (ATA), when the scattering matrix of the individual atoms in a random system is replaced by an average t-matrix.

After a straightforward algebra one gets for the density of electrons of E energy, inside a muffin-tin sphere

$$g(\underline{r},E) = \frac{\varkappa}{4\pi} \sum_{L_1L_2} R_{\ell_1}(r)R_{\ell_2}(r)Y_{L_1}(\hat{r})Y_{L_2}(\hat{r}) * \left[\delta_{L_1L_2} - \mathrm{Im}\, \frac{1}{\varkappa} \sum_{L} J^{-1}_{L_1L}\, M_{LL_2} \right] \tag{3}$$

Here $R_\ell(r)Y_L(\hat{r})$ is the regular solution of the radial Schrödinger equation with the muffin-tin potential and for the energy $E = \varkappa^2$. The matrices J and M are defined as follows:

$$J_{L_1L_2} = [1-tD]_{L_1L_2}, \tag{4}$$

$$[tD]_{L_1L_2} = t_{\ell_2} \cdot \sum_{\underline{R}\neq o} B_{L_1L_2}(\underline{R},\varkappa), \tag{5}$$

$$M_{L_1L_2} = \sum_{L'} t_{\ell'} \sum_{R\neq o} B_{L_1L'}(-\underline{R},\varkappa) B_{L'L_2}(\underline{R},\varkappa), \tag{6}$$

where $t_\ell = t_\ell(\varkappa)$ and $B_{L_1L_2}$ are the expansion cofficients of the t-matrix and the free-particle Green's functions, respectively, defined in the terms of phase shifts, $\eta_\ell(E)$, and of the spherical Hankel functions, $h^+_\ell(\varkappa R)$ in the following way:

$$t_\ell = \frac{1}{\varkappa} \sin \eta_\ell(E) e^{i\eta_\ell(E)}, \tag{7}$$

$$B_{L_1L_2}(\underline{R},\varkappa) = -\, 4\pi i\varkappa \sum_{L'} i^{(\ell_1-\ell_2-\ell')} C_{L_1L_2L'} Y_{L'}(R) h^+_{\ell'}(\varkappa R). \tag{8}$$

Equation (3) gives the density of electrons only for a given configuration of the atoms, represented by their coordinates, $\underline{R}_i$. To calculate the ensemble average of $g(\underline{r},E)$ one has to make some assumption for the distribution of atoms in the system, then to define a decoupling scheme, how to calculate the many-particle averages in equation (3). A particularly simple formula is resulted if one supposes that the atomic distributions can be represented by a spherically symmetric pair distribution function, g(R) and if, for the higher order terms, one applies Kirkwood's decoupling schemes. In that case the sum in the (tD) and (M) matrices, over the atomic coordinates, can be replaced by integrals containig the function g(R). Due to the isotropy of this distribution the above mentioned matrices are diagonal ones, and the inverse of J can easily be calculated. Carrying out the integrations with respect to the angular coordinates one gets the very simple final formulas for this case as follows:

$$\rho(r,E) = \frac{\varkappa}{\pi} \sum_{\ell} \frac{2\ell+1}{4\pi} R_{\ell}^{2}(r) \left[1-\mathrm{Im}\, \frac{1}{\varkappa} J_{\ell}^{-1} M_{\ell}\right] \tag{9}$$

$$J_{\ell} = [1-tD]_{\ell}, \quad [tD]_{\ell} = -4\pi\rho_{o} t_{\ell} I \tag{10}$$

$$M_{\ell} = -(4\pi\varkappa)^{2} \sum_{\ell',\ell''} t_{\ell'} D(\ell,\ell';\ell'') H_{\ell''} \tag{11}$$

where ρ_o is the average density of the material, the simple numbers $D(\ell,\ell';\ell'')$ can easily be calculated from the Clebsch-Gordon coefficients, and the I and H_{ℓ} are integrals:

$$I = \int_{0}^{\infty} e^{i\varkappa R} g(R) R dR, \tag{12}$$

$$H_{\ell} = \int_{0}^{\infty} h_{e}^{+2}(\varkappa R) g(R) R^{2} dR. \tag{13}$$

The integral of $\rho(r,E)$ inside the atomic volume with respect to the coordinates $\underline{r}$ leads to the local density of states, $\rho(E)$.

Preliminary numerical calculations were performed for liquid or amorphous iron. The $g(R)$ pair distribution function was calculated from the analitical structure factor of the hard sphere solution of the Percus-Yevick equations. The muffin-tin potential was given by a simple analytical expression, the only parameter of it was choosen to reproduce the resonance in the $\eta_2(E)$ phase-shift. Using these ingredients a single-peaked local density of states curve was resulted *(Fig. 1.)*, which, in our model, corresponds to a single scattering center embedded into a medium. The effect of this medium is expressed by the diagonal matrices J_{ℓ} and M_{ℓ}, and resulted in the broadening of the resonance level characteristic for transitional metals.

To improve the model described we intend to take into account the local environment of the individual atoms by dividing the sums over $\underline{R}_i$ in formulas (5) and (6) into two parts. In the first one the $\underline{R}_i$ takes the values of the few neighbours around the original atom, creating a cluster, in the second parts $\underline{R}_i$ runs over the atomic coordinates outside of this cluster. Also, the model muffin-tin potential will be replaced by a more realistic one, calculated from the wave functions of the neighbouring atoms. This sort of calculations are in progress and will be reported in later publications.

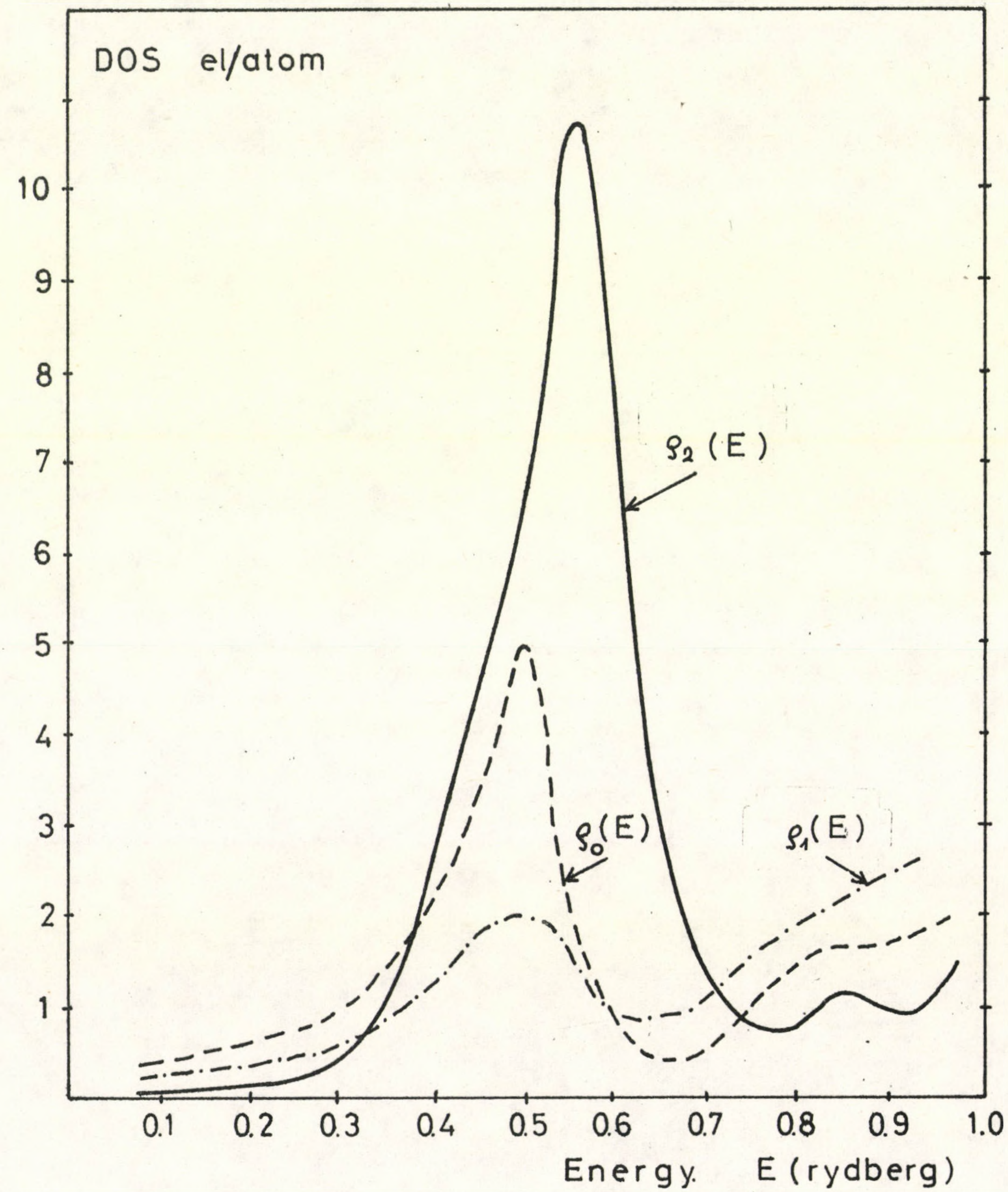

Fig. 1. Local density of states for amorphous Fe

REFERENCE

[1] L. Schwartz, H. Ehrenreich: Annals of Physics 64, 100-148 (1971);

L. Schwartz, H.K. Peterson, A. Bansil: Phys. Rev. B12, 3113-3123 (1975);

J. Bethell, J.L. Beeby: J. Phys. F: Metal Phys. 7, 1193-1205 (1977);

S. Asano, F. Yonezawa: J. Phys. F: Metal Phys. 10, 75-79 (1980).

Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Tompa Kálmán
Szakmai lektor: Hargitai Csaba
Nyelvi lektor: Hargitai Csaba
Gépelte: Végvári Istvánné
Példányszám: 220 Törzsszám: 80-633
Készült a KFKI sokszorosító üzemében
Felelős vezető: Nagy Károly
Budapest, 1980. október hó