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Electron structure of topologically disordered metals

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Here two methods for calculating the density of states of electrons in conduction band of disordered metals are investigated. The first one is based on the usage of one-parameter trial electron wave function. The equation for density of states gotten within this method is more general as compared to the results of perturbation theory. Electron-ion interaction is applied in the form of electron-ion structure factor, which makes it possible to use this method for a series of systems where potential form factor is not a small value and the perturbation theory fails. It also gives us well-known results of Relel-Schrodinger and Brilliuen-Vigner perturbation theory in case of small potential. Basically, the second approach is a common perturbation theory for pseudo-potential and Green's function method. It considers the contributions up to the third order. The results of computation for density of states in some non-transition metals are presented. The deviation of density of states causing the appearance of pseudo-gap is clearly recognized.

Key words: *density of states, mass operator, form factor, structure factor, pseudo-gap*

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Introduction

Disorder metallic systems, including liquid, amorphous metals and metallic glasses are still investigated intensively due to the prospects of their industrial use. Some unique properties of such systems cannot be explained within crystalline physics methods. The problem of disorder remains to be one of the most urgent and has not been ultimately solved in condensed matter physics. Disordered state differs from crystalline one by the absence of long range ordering and makes it impossible to use the translation symmetry methods [1]. Nonequilibrium of the structure makes us apply the configuration averaging over all accidentally occupied ion locations [2].

Accented features of disordered metals bring about a number of anomalous characteristics for many thermodynamic properties. One of the most interesting problems of disordered condensed matter physics for quantum systems is the description of the

formation of pseudo-gap in the density of electron state. Two methods of calculating these characteristics are presented below.

1. Variation approach

Another method for calculation of density of states is the variation one. The calculation scheme for this method was proposed in [3]. Setting the wave function and energy of the ground state of an electron in disordered metal media, where $\mathbf{R_i}$ is the i^{th} ion coordinate, as the known values ϕ_0 and E_0 respectively, we assume the wave function of the excited state

$$\psi_k = \chi_k \psi_0 \,, \tag{1.1}$$

where

$$\chi_k = \frac{1}{\sqrt{C_k}} \mathrm{e}^{\mathbf{ikr}} \left\{ 1 + \frac{1}{\sqrt{N}} \sum_{\mathbf{q}\neq\mathbf{0}} u_k(q) \rho_{\mathbf{q}} \mathrm{e}^{\mathbf{iqr}} \right\} \tag{1.2}$$

and

$$\rho_{\mathbf{q}} = \frac{1}{\sqrt{N}} \sum_j \mathrm{e}^{\mathbf{iqR_i}} \,.$$

It corresponds to an eigenstate of both full impulse operator for eigenvalue $\hbar k$ and Hamiltonian – E_k. Obviously, we have the one parameter trial function for Ritz's condition of the variation problem for energy spectrum of an electron in the conduction band. We use the following condition to define C_k

$$C_k = |\chi_k|^2 \,. \tag{1.3}$$

Now, the variation problem transforms into the next functional equation

$$E_k = E_0 + \frac{\hbar^2 \langle |\nabla \chi_k|^2 \rangle}{2m} \,. \tag{1.4}$$

Calculating the energy spectrum of the electrons in the conduction band, we consider the one-sum contributions and neglect many-particle correlations. In this case, we get minimum magnitude of Ritz's functional in case

$$u_k(q) = -\frac{S_{\mathrm{ei}}(q)\{E_k - \frac{\hbar^2}{2m}\mathbf{k}(\mathbf{k}+\mathbf{q})\}}{S_q\{E_k - \frac{\hbar^2}{2m}(\mathbf{k}+\mathbf{q})^{\mathbf{2}}\}} \,. \tag{1.5}$$

Here $S_q = \langle \rho_{\mathbf{q}} \rho_{-\mathbf{q}} \rangle$ – is a structure factor of disordered metal and $S_{\mathrm{ei}}(k,q) = \langle \rho_{\mathbf{q}} \mathrm{e}^{-\mathbf{ikr}} \rangle$ – electron-ion structure factor of the system. The latter can be presented in Born approximation in case of small potential [4] as

$$S_{\mathrm{ei}}(k,q) = -\frac{2S_q \langle k+q|w|q \rangle}{\hbar^2 q^2/2m} \,. \tag{1.6}$$

Applying this result to equations (1.1)–(1.4) and substituting $E_k = \hbar^2/2m(k^2 + \Delta_k)$ we define

$$\Delta_k = \frac{\Omega_0}{8\pi^2 I_k}\int_0^\infty \mathrm{d}qq^2\frac{S_{\mathrm{ei}}(k,q)^2}{S_q}\left\{q^2 - \frac{q^4-\Delta_k^2}{4kq}\ln\frac{|q^2-\Delta_k+2kq|}{|q^2-\Delta_k-2kq|} + \frac{\Delta_k(q^2+\Delta_k)^2}{(q^2-\Delta_k)^2-4k^2q^2}\right\}. \quad (1.7)$$

Here

$$I_k = 1 + \frac{\Omega_0}{8\pi^2}\int_0^\infty \mathrm{d}qq^2\frac{S_{\mathrm{ei}}(k,q)^2}{S_q}\left\{-3 + \frac{q^2+\Delta_k}{2kq}\ln\frac{|q^2-\Delta_k+2kq|}{|q^2-\Delta_k-2kq|} + \frac{(q^2+\Delta_k)^2}{(q^2-\Delta_k)^2-4k^2q^2}\right\}. \quad (1.8)$$

This result can be easily transformed into Relel-Schrodinger theory expression [6] for energy spectrum by assuming $\Delta_k = 0$ and $I_k = 1$. Moreover, it can also be transformed by the same assumption, but saving $\Delta_k \neq 0$ in logarithm, to Brilliuen-Vigner theory result [6]. Thus, for density of states reduced by its free-electron approximation $N_0(E) = \Omega_0 k/\pi^2$ we have

$$g(E) = \frac{N(E)}{N_0(E)} = \left(1 + \frac{\Omega_0}{16\pi^2k^2I_k}\int_0^\infty \mathrm{d}qq^2\frac{S_{\mathrm{ei}}(k,q)^2}{S_q} \times\left\{\frac{(q^2+\Delta_k)^2}{2kq}\ln\frac{|q^2-\Delta_k+2kq|}{|q^2-\Delta_k-2kq|} - \frac{(q^2-\Delta_k)(q^2+\Delta_k)^2}{(q^2-\Delta_k)^2-4k^2q^2}\right\}\right)^{-1}. \quad (1.9)$$

This expression consequently follows the expression (1.7)–(1.8) if density of states is assumed

$$N(E) = \frac{\Omega_0 k^2}{\pi^2}\frac{\mathrm{d}k}{\mathrm{d}E}.$$

The calculations of density of states within this theory were presented in [7]. The pseudo-gap deviation of density of states in close range of Ferme level calculated in Relel-Schrodinger and Brilliuen-Vigner approximation of perturbation theory was obvious and similar. Both were in good correlation with Zaiman's and Mott's predictions for distribution of electrons in disordered metals. Variation approach used in [7] gives a better agreement than the results of second order of perturbation theory in terms of electron-ionic potential.

2. Perturbation theory

The well-known fact is that configuration averaging must be used in disordered systems [1]. So, when expressing the electron Green's function averaged over configurations as

$$G(E,k)_{\mathrm{conf}} = \frac{1}{E - \frac{\hbar^2k^2}{2m} - \Sigma(E,k)_{\mathrm{conf}}} \quad (2.1)$$

we achieve the following form for the mass operator in the third order of perturbation theory [5]

$$\begin{aligned}\Sigma(E,k)_{\rm conf} &= \langle \mathbf{k}|\mathbf{W}|\mathbf{k}\rangle + \frac{\mathbf{1}}{\mathbf{N}}\sum_{\mathbf{q}\neq\mathbf{0}} \mathbf{S_q}\frac{\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}\rangle}{\mathbf{E}-\hbar^{\mathbf{2}}/\mathbf{2m}(\mathbf{k}+\mathbf{q})^{\mathbf{2}}} \\ &+ \frac{1}{\sqrt{N}^3}\sum_{q'\neq 0}\sum_{q\neq q'\neq 0} S_{q,q',q-q'}\frac{\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}'\rangle\langle \mathbf{k}+\mathbf{q}'|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}\rangle}{[E-\hbar^2/2m(\mathbf{k}+\mathbf{q}')^{\mathbf{2}}][\mathbf{E}-\hbar^{\mathbf{2}}/\mathbf{2m}(\mathbf{k}+\mathbf{q})^{\mathbf{2}}]} \\ &+ \frac{1}{N}\sum_{q\neq 0} S_q\frac{\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}\rangle}{[E-\hbar^2/2m(\mathbf{k}+\mathbf{q})^{\mathbf{2}}]^{\mathbf{2}}}\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle \qquad (2.2)\end{aligned}$$

and this can be easily shown by factoring the Green's function into a row for pseudopotential. Here we take the following designations: $W(r)=\sum_i w(\mathbf{r}-\mathbf{R_i})$ – electron-ion potential.

Picking out imaginary and real parts of Green's function we get for real contribution of the first and second order

$$\begin{aligned}\Sigma'^{(2)}(E,k)_{\rm conf} &= \langle \mathbf{k}|\mathbf{w}|\mathbf{k}\rangle_{\theta,\varphi} - \frac{\mathbf{\Omega_0}}{\mathbf{4\pi^2}}\frac{\mathbf{m}}{\hbar^{\mathbf{2}}}\int_0^\infty \mathrm{d}\mathbf{q}\mathbf{q}/\mathbf{k}\mathbf{S_q}|\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|^{\mathbf{2}} \\ &\times \ln\frac{|E-\hbar^2/2m(k+q)^2|}{|E-\hbar^2/2m(k-q)^2|} \qquad (2.3)\end{aligned}$$

and for the imaginary one

$$\begin{aligned}\Sigma''^{(2)}(E,k)_{\rm conf} &= \frac{\Omega_0}{4\pi^2}\frac{m}{\hbar^2}\int_0^\infty \mathrm{d}qq/kS_q|\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|^{\mathbf{2}} \\ &\times \int_{-1}^{1}\mathrm{d}x\delta\left\{E-\hbar^2/2m(k^2+q^2+2kqx)\right\} \\ &= \frac{\Omega_0}{4\pi^2}\frac{m}{\hbar^2}\int_{k-\sqrt{2mE}/\hbar}^{k+\sqrt{2mE}/\hbar}\mathrm{d}qq/kS_q|\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|^{\mathbf{2}}\,. \qquad (2.4)\end{aligned}$$

Now, consider the third order correction. We set the convolution approximation for three-particle structure factor $S_{q,q',q-q'}=S_qS_{q'}S_{|\mathbf{q}-\mathbf{q}'|}$ [1]. The third order contribution takes the form of 2 last items in (1.2); then for real part of this correction we have

$$\begin{aligned}\Sigma'^{(3)}(E,k)_{\rm conf} &= \frac{\Omega_0^2}{(2\pi)^4}\frac{m^2}{\hbar^4}\int_0^\infty \mathrm{d}qq/k\int_0^\infty \mathrm{d}q'q'/kS_qS_{q'}S_{|\mathbf{q}-\mathbf{q}'|} \\ &\times \langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}'\rangle_{\theta,\varphi}\langle \mathbf{k}+\mathbf{q}'|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}\rangle_{\theta,\varphi}\ln\frac{|\mathbf{E}-\hbar^{\mathbf{2}}/\mathbf{2m}(\mathbf{k}+\mathbf{q})^{\mathbf{2}}|}{|\mathbf{E}-\hbar^{\mathbf{2}}/\mathbf{2m}(\mathbf{k}-\mathbf{q})^{\mathbf{2}}|} \\ &\times \ln\frac{|E-\hbar^2/2m(k+q')^2|}{|E-\hbar^2/2m(k-q')^2|} + \frac{2\Omega_0}{4\pi^2}\frac{m}{\hbar^2}\int_0^\infty \mathrm{d}qq/kS_q\langle \mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi} \\ &\times \frac{|\langle \mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|^{\mathbf{2}}}{|E-\hbar^2/2m(k+q)^2||E-\hbar^2/2m(k-q)^2|}\,. \qquad (2.5)\end{aligned}$$

Finding an imaginary part, we had better consider those two items separately and using the calculation method similar to the second order calculation for imaginary contribution and taking into account the following equation for transformation of the divisor

$$\frac{1}{(E-\hbar^2/2mq^2)(E-\hbar^2/2mq'^2)}=\frac{1}{\hbar^2/2m(q^2-q'^2)}\left\{\frac{1}{E-\hbar^2/2mq^2}-\frac{1}{E-\hbar^2/2mq'^2}\right\}$$

we get for the first item

$$\Sigma_I''^{(3)}(E,k)_{\rm conf}=\frac{\Omega_0}{4\pi^2}\frac{m}{\hbar^2}\int_{k-\sqrt{2mE}/\hbar}^{k+\sqrt{2mE}/\hbar}{\rm d}qq/k\int_0^\infty {\rm d}q'q'/kS_qS_{q'}S_{|\mathbf{q}-\mathbf{q}'|}\langle\mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}'\rangle_{\theta,\varphi}$$
$$\times\langle\mathbf{k}+\mathbf{q}'|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}\langle\mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}\rangle_{\theta,\varphi}\ln\frac{|\mathbf{E}-\hbar^\mathbf{2}/\mathbf{2m}(\mathbf{k}+\mathbf{q}')^\mathbf{2}|}{|\mathbf{E}-\hbar^\mathbf{2}/\mathbf{2m}(\mathbf{k}-\mathbf{q}')^\mathbf{2}|} \quad (2.6)$$

and the following expression for the second item

$$\begin{aligned}\Sigma_{II}''^{(3)}(E,k)_{\rm conf} &= \frac{2\Omega_0}{4\pi^2}\frac{m}{\hbar^2}\int_0^\infty {\rm d}qq/kS_q\langle\mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|\langle\mathbf{k}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}|^\mathbf{2}\\ &\quad\times\left\{\delta(E-\hbar^2/2m(k+q)^2)-\delta(E-\hbar^2/2m(k-q)^2)\right\}\\ &= \frac{\Omega_0}{4\pi^2}\frac{m}{\hbar^2}S_q\langle\mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}\times\frac{\sqrt{\mathbf{2mE}}-\mathbf{k}}{\mathbf{k}\sqrt{\mathbf{2mE}}}_{\mathbf{q}=|\mathbf{k}-\sqrt{\mathbf{2mE}}|}\\ &\quad-\frac{\Omega_0}{4\pi^2}\frac{m}{\hbar^2}S_q\langle\mathbf{k}+\mathbf{q}|\mathbf{w}|\mathbf{k}+\mathbf{q}\rangle_{\theta,\varphi}\times\frac{\sqrt{\mathbf{2mE}}+\mathbf{k}}{\mathbf{k}\sqrt{\mathbf{2mE}}}_{\mathbf{q}=\mathbf{k}+\sqrt{\mathbf{2mE}}}. \quad (2.7)\end{aligned}$$

Density of states of electrons in conduction band is defined as the imaginary part of the electron Green's function averaged over all possible configurations

$$\begin{aligned}N(E) &= \frac{1}{\pi}{\rm Sp}\,{\rm Im}\,G(E-{\rm i}\varepsilon,k)_{\rm conf}\\ &= \frac{1}{\pi}\sum_{\mathbf{k}}\frac{\Sigma''(E,k)_{\rm conf}}{[E-\frac{\hbar^2k^2}{2m}-\Sigma'(E,k)_{\rm conf}]^2+\Sigma''(E,k)^2_{\rm conf}}. \quad (2.8)\end{aligned}$$

Here

$$\begin{aligned}\Sigma'(E,k)_{\rm conf} &= \Sigma'^{(2)}(E,k)_{\rm conf}+\Sigma'^{(3)}(E,k)_{\rm conf},\\ \Sigma''(E,k)_{\rm conf} &= \Sigma''^{(2)}(E,k)_{\rm conf}+\Sigma''^{(3)_I}(E,k)_{\rm conf}+\Sigma''^{(3)_{II}}(E,k)_{\rm conf}.\end{aligned}$$

Proceeding to integral over impulse k and taking into account spin degeneracy factor 2, we finally get the following expression for density of states.

$$N(E)=\frac{\Omega_0}{\pi^3}\int_0^\infty\frac{\Sigma''(E,k)_{\rm conf}k^2{\rm d}k}{[E-\frac{\hbar^2k^2}{2m}-\Sigma'(E,k)_{\rm conf}]^2+\Sigma''(E,k)^2_{\rm conf}}. \quad (2.9)$$

Some computation results are presented below. Form factor of electron-ion interaction is calculated for the following non-local model potential

$$W^{(0)}(r) = -\frac{Z}{r} + \sum_l \mathrm{e}^{-\frac{r}{R_l}} \left(\frac{Z}{r} + A_l\right) P_l \,, \tag{2.10}$$

here A_l, R_l – are model parameters of potential, P_l – is the projecting operator.

Screened form factor of this potential takes the following form [5]

$$w(q) = -\frac{4\pi Z}{q^2 \Omega_0 \epsilon^*(q)} + f(k,q) - \left(1 - \frac{\varphi(q)}{\epsilon^*(q)}\right) \frac{4}{\pi q^3 \epsilon(q)} \int_0^{k_\mathrm{F}} f(k,q) \ln \left|\frac{k-2q}{k+2q}\right| k\mathrm{d}k. \tag{2.11}$$

Here

$$f(k,q) = \sum_{l=0}^{l_0} \frac{4\pi(2l+1)}{\Omega_0} P_l(\cos\theta) \int_0^\infty \mathrm{e}^{-\frac{r}{R_l}} \left(\frac{Z}{r} + A_l\right) j_l(kr) j_l\left(|k^2+q^2|r\right) r^2 \mathrm{d}r \tag{2.12}$$

is non-local part of form factor and

$$\epsilon(q)^* = 1 - (1 - \varphi(q))(\epsilon(q) - 1) \tag{2.13}$$

is a dielectric permittivity accounting for the processes of electron exchange and correlation. Here we used Hartry approximation for permittivity $\epsilon(q)$ and Heldart-Vosko approximation for local field correction $\varphi(q)$.

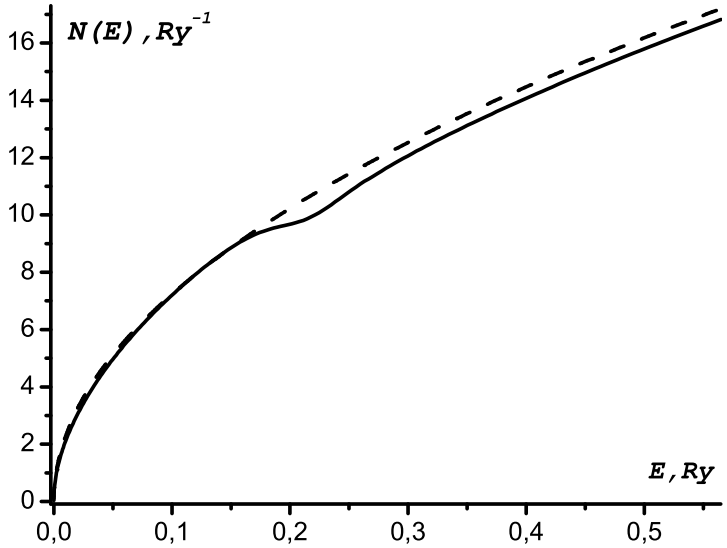

Figure 1. Density of states Cd-II.

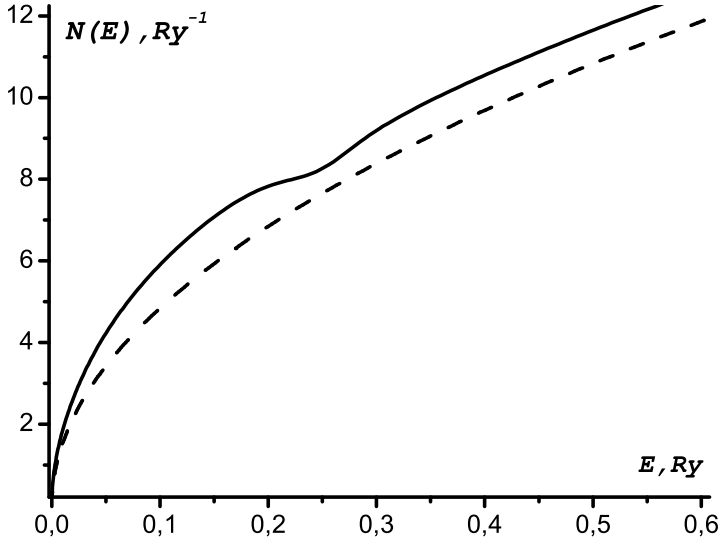

Figure 2. Density of states Zn-II.

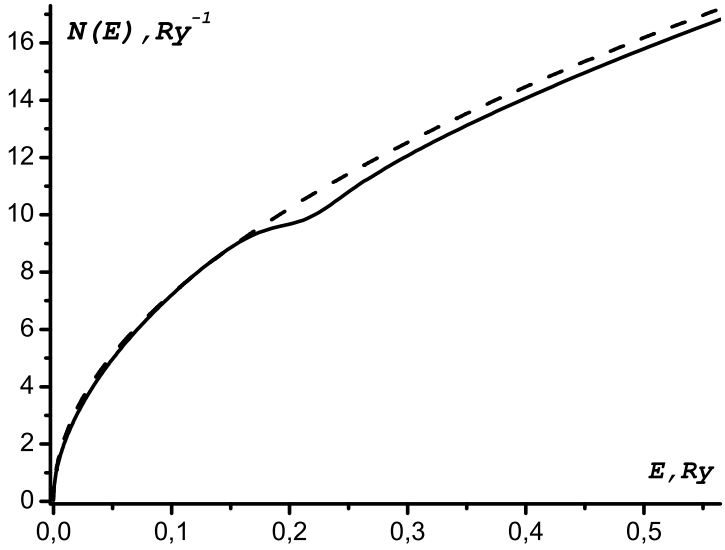

Figure 3. Density of states Al-III.

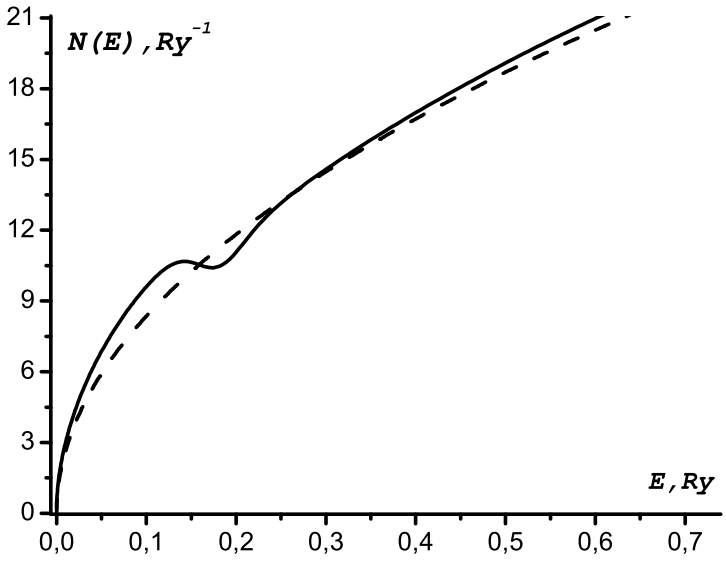

Figure 4. Density of states In-III.

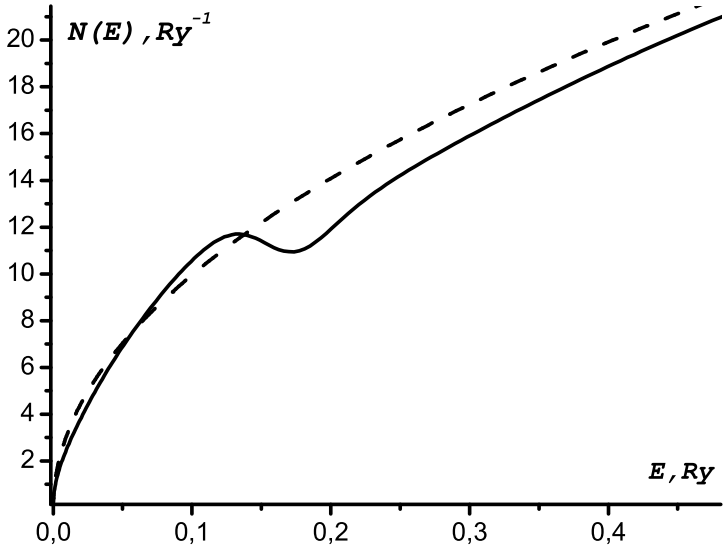

Figure 5. Density of states Pb-IV.

So, in figures 1–6 we indicate the perturbation theory result with solid line and free electron approximation result with dash line. We use Ashkroft-Lekner result of Percus-Yevik approximation for structure factor in our calculations.

Evidently, electron density of states has a pseudo-gap behavior near Ferme level range. In approximation not responsive to the imaginary part of mass operator the second order gap takes place and in case of this theory the imaginary part of mass operator spreads this gap and transforms it into the pseudo-gap deviation of density of states. This happens due to the extinction of electron spectrum on structure fluctuations.

We have also investigated the effects of the order of perturbation theory during these calculations. For some metals (Cd,In,Pb) the third order contribution is the most determinant and for the others (Zn,Al) the second order is already good approximation for density of states.

The calculations within the non-local pseudopotential theory are more accurate than the calculations within its local approximation (i. e., Ferme sphere approximation)and makes it possible to exactly define the shape and position of pseudo-gap minimum on density of states distribution.

3. Conclusion

Our results show that in order to increase the accuracy of data on density of states of disordered metals we must provide the non-local properties of pseudopotential and the higher orders of perturbation theory in our calculations.

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Електронна структура топологічно невпорядкованих металів

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В даній роботі розглядаються два різних підходи до визначення густини станів та енергетичного спектру електронів провідності у невпорядкованих металах. Перший з них грунтується на варіаційному принципі з використанням однопараметричної пробної хвильової функції електронів провідності. Для енергетичного спектру отримано рівняння, яке має більш загальний вигляд у порівнянні з результатами теорії збурень. Електрон-іонна взаємодія входить в теорію через електрон-іонний структурний фактор, що дає змогу застосувати теорію і в тих випадках, коли формфактор потенціалу не є малою величиною і теорія збурень не може бути застосована. Якщо формфактори екранованого потенціалу є малими, то із виведеного варіаційного виразу в часткових випадках отримуються відомі результати теорії збурень Релея-Шредінгера та Бріллюена-Вігнера. Другий підхід пов'язаний з використанням методу функцій Гріна та стандартної теорії збурень за псевдопотенціалом з урахуванням членів до третього порядку включно. Для ряду металів виконані чисельні розрахунки густини електронних станів. Виявлено помітне відхилення відносно вільноелектронного наближення на залежностях густини станів від енергії, що обумовлює появу псевдощілини.

Ключові слова: *густина станів, масовий оператор, формфактор, структурний фактор, псевдощілина*

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