

Changing the properties of metals under conditions of exposure to ionizing radiation

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Abstract. The equation state for metals were provided on the basis of the pseudopotential method when part of the ionic cores further ionized. The isotherms were obtained for different degrees of the atoms ionization on the example of aluminium. The change in the physical properties of metals were analyzed under conditions of excitation by ionizing radiation of the electron subsystem. The interatomic interaction potentials were calculated for the case of a local increase concentration of conduction electrons.

1. Introduction

While studying the radiation-stimulated processes it is necessary to learn out not only the interatomic coupling potential, but the consistent relations considering the ionized particles, as well. Such relations can be provided on the basis of binding energy.

According to the pseudopotential, the binding energy in the model of the second order looks as follows:

$$E(\Omega) = E_e^{(0)} + E_e^{(1)} + E_e^{(2)} + E_i. \quad (1)$$

Where Ω -atomic volume, $E_e^{(0)}$ -the energy of homogeneous electron gas, including the kinetic, exchange and correlative energy of conduction electrons, $E_e^{(1)}$ -the energy, caused by the fact that the ions are not the points and have some dimensions, $E_e^{(2)}$ -energy of zonal structure, E_i -Evald energy.

The registration of differently ionized atoms was being made on the base of technique, been developed for the alloys of replacement [1].

The lattice is placed in homogenous negative charge with density \bar{Z}/Ω in concordance with the technique.

Here

$$\bar{Z} = Z_3 C + Z_4 (1 - C), \quad (2)$$

Z_3 is the charge of three-, Z_4 is the four – charged ions, C is the concentration of Z_3 ions.

The calculations was being defined assuming that δ -electrons were leaving the observed area, that is $\bar{Z} = 3$.

The energy of homogeneous electron gas per rydberg on electron has a form [2]:

$$E_e^{(0)} = Z \left(\frac{2,21}{r_s^2} - \frac{0,916}{r_s} + E_c \right), \text{Ry} \quad (3)$$

Here $r_s = (3/4\pi n_0)^{1/3}$ is the usual radius giving the volume per electron, $n_0 = \bar{Z}/\Omega$, E_c is the correlation energy. Expression for E_c may be written as Nozieres – Pines approximation [2]:



$$E_c = -0,115 + 0,031 \ln(r_s). \quad (4)$$

The energy connected to the not Coulomb part of the potential has a form [3]:

$$E_e^{(1)} = 2 \left(\frac{3 Z \bar{Z}}{2 R_a} \left(\frac{R_m}{R_a} \right)^2 - \bar{A} \bar{Z} \left(\frac{R_m}{R_a} \right)^3 \right), \text{ Ry} \quad (5)$$

\bar{A} is the average level between A_i of the potential well.

The energy of zonal structure looks as follows:

$$E_e^{(2)} = -\Omega \sum_{\vec{\tau}} \frac{\pi(\vec{\tau})}{\varepsilon(\vec{\tau})} |V(\vec{\tau})|^2, \quad (6)$$

where $\vec{\tau}$ is the reciprocal lattice vector. Here the vector $\vec{\tau} = 0$ is absent.

The static dielectric permeability of the conduction electrons is written as:

$$\varepsilon(q) = 1 + \frac{4\pi}{q^2} \pi(q), \quad (7)$$

where q is the wave vector.

The polarization operator has a form:

$$\pi(q) = \frac{\pi_0(q)}{1 - 4\pi(G(q)/q^2)\pi_0(q)}. \quad (8)$$

Hartre permittivity it is:

$$\pi_0(q) = \frac{3 n_0}{2 \varepsilon_F} \left[\frac{1}{2} + \frac{1-x^2}{4x} \ln \left| \frac{1+x}{1-x} \right| \right], \quad (9)$$

where $x = q/2k_F$, n_0 is the concentration of the conduction electrons.

Ewald energy has a form:

$$E_i = -\frac{\bar{Z}^2 e^2}{r_0} a_M, \quad (11)$$

where $r_0 = (3\Omega / 4\pi)^{1/3}$, a_M is Madelung constant, $a_M = 1.7917$ for face-centered cubic lattice.

Static component of pressure- $P_{st}(\Omega)$ can be defined by equation

$$P_{st} = -\frac{\partial E}{\partial \Omega}. \quad (12)$$

Differentiation E on volume Ω was made under the formula:

$$\frac{\partial}{\partial \Omega} = \left(\frac{\partial}{\partial \Omega} \right)_{n_0, \tau, \mathbf{R}} + \frac{\partial \mathbf{R}}{\partial \Omega} \left(\frac{\partial}{\partial \mathbf{R}} \right)_{n_0, \Omega, \tau} + \frac{\partial \tau}{\partial \Omega} \left(\frac{\partial}{\partial \tau} \right)_{n_0, \Omega, \mathbf{R}} + \frac{\partial n_0}{\partial \Omega} \left(\frac{\partial}{\partial n_0} \right)_{\tau, \Omega, \mathbf{R}} \quad (13)$$

The energy is function of volume. The volume depends from n_0 , $k_F = (3\pi^2 n_0)^{1/3}$, r_s , $\mathbf{R} = a\mathbf{m}$, $\tau = \mathbf{n}2\pi/a$, where a is the lattice constant, \mathbf{R} is lattice vector.

The contribution caused by harmonic phonon pressure and zero oscillation energy has been negligible.

The $E_e^{(0)}$ was defined assuming that δ -electrons were leaving the observed area and the concentration of conductivity electrons was constant [4].

Calculating the $E_e^{(1)}$ and $E_e^{(2)}$, the model potential parameters (after quantum defect method [4]) had been taken. Figure 1 shows the aluminium isotherms at the 0 °K temperature for the different grades of ionization. So the 10% ionization results in the 15 kbar internal pressure appearance.

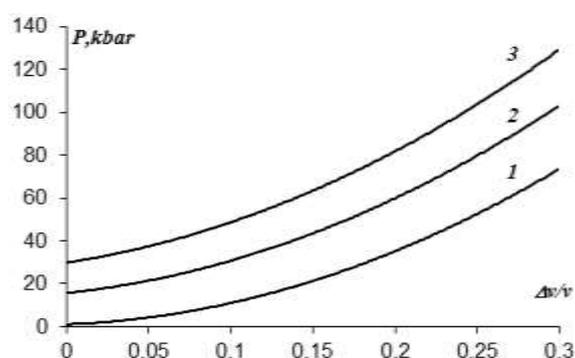


Figure 1. Aluminum isotherms for 0 K temperature: 1 – par, 2 – 10% of ionized atoms; 3 – 20% of ionized atoms.

Thus, the equation of the matter states for the case of ionization were investigated by means of the given method.

The consistent relations, resultant, are proposed to be engaged for studying the destruction of surfaces under the charged particles sturdy beam bombardment.

It is known that intensive radiation generates ionized atoms within the track of a fast charged particle as well as near the solid surface. The potentials of interactions between these atoms and surrounding particles change considerably. The knowledge of corresponding potentials of interatomic interaction is necessary for studying the behavior of partially or entirely ionized matter.

2. Change of Interatomic Forces upon Excitation of Electronic Subsystem

Essential part of energy of fast charged particle in metal is spent on ionization of atoms and excitation of conduction electrons. Electrons accelerating in the field of charged particle slow down transferring their energy to conduction electrons in the space limited by the radius of about 10 interatomic distances from track. For example, density of electronic energy reaches 10^{12} erg/cm³ [5] in the area of fission fragment particle trajectory, which corresponds to approximately double increase in the energy of free electrons in aluminum over that of Fermi.

Though the fact of change of the interatomic interaction nature upon excitation of electronic subsystem does not raise any doubt, the experimental observation of it is quite a task. In paper [6] there is an analysis of this phenomenon on the basis of experimental data on the threshold energy of the atom displacement from the equilibrium state E_d , which is determined by change of the value of some physical quantity, which is dependent on concentration of point defects (electric conductance for example) under changing energy of falling particles.

The displacement threshold was determined in many papers. In different experiments different values were obtained for one and the same elements even for polycrystalline materials. Comparison of experimental results indicates the existence of some dependence of E_d value on the interval of energies of bombarding particles.

The analysis of work enumerated in [6], enable to conclude that the lower the applied down limit of bombarding particle energy (0.1 – 3 MeV in the considered energy range), the smaller threshold energy of displacement. Peculiarities of E_d behavior depending on the energy of incident particle can be explained by change of the nature of interatomic interaction in metal under conditions of excitation of electronic subsystem. During movement the fast charged particle loses energy transferring it to free electrons and to electrons of the ion core, and in case of collision with the core ion – directly to it. It can be assumed that excitation wave moves in front of the accelerated particle. Thus, already before the collision with the fast charged particle, the initially knocked-on atom and surrounding ions are in the state other than equilibrium.

In order to find out whether the existence of such area is possible let us consider the times of energy re-distribution in metal. According to [7], the characteristic time of excitation relaxation of a separate electron is about 10^{-15} s, and characteristic time of excitation transfer from the electron gas to the ion subsystem is 10^{-12} s (characteristic time of the lattice temperature change from the moment of fast charged particle track occurrence associated with the energy exchange between Fermi electrons and ion cores). Time of equilibration between the electron gas and the lattice is about 10^{-12} s. Thus, if the period of atom transition from one equilibrium state to another is inversely proportional to Debye frequency which is about 10^{-13} s, then quite a large number of transitions of ions amid the electrons with enhanced energy is possible during the time of excitation state existence.

In figure 2 the displacement threshold energies obtained by extrapolation taken from paper [6] are presented in comparison with the graphs of primary electrons energy losses for ionization and excitation of free electron gas

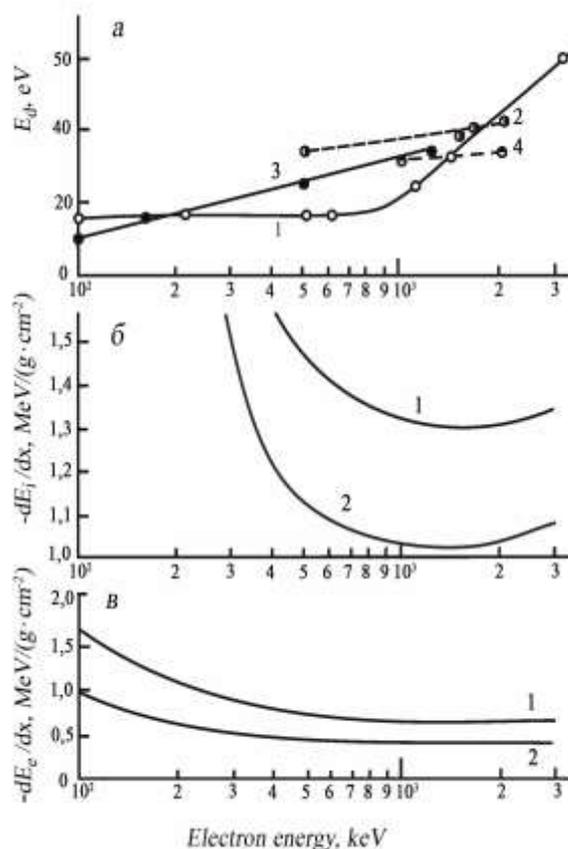


Figure 2. Parameters of interaction of radiation with metals depending on energy of bombarding electrons: a – threshold displacement energy; б – energy losses for initial ionization; B – energy losses for electronic excitation: 1- Al; 2 – W; 3 – Cu; 4 – Mo..

In experiments for determination of displacement threshold the target thickness is much smaller than the length of electron free path, therefore the energy absorbed by electrons is proportional to dE/dx . As is seen from the figure, the correlation between losses for electronic slow down and the value of displacement threshold is observed. The more energy is transferred to the electronic subsystem, the lower is experimentally observed value of the displacement threshold. The effect of the

displacement threshold dependence on the energy introduced to the electronic subsystem is also observed upon investigation of monocrystalline samples depending on the crystallographic direction.

Phenomena associated with non-elastic sputtering of metals by ion bombardment are examined in paper [8]. The local heating of electronic subsystem which results in interatomic interaction forces change was considered as one of the reasons for abnormally high fission fragments sputtering factor.

Besides, it was discovered experimentally that gamma and electron radiation of comparably weak intensity have an essential influence on phase conversions in metals and alloys [9]. In particular, under the influence of gamma radiation the ordering temperature of β - Cu - Zn alloy is shifted. A hypothesis was stated that gamma radiation causes “softening” of the crystalline lattice, i.e. the decrease in elastic moduli which is manifested in the ordering temperature shift. Since the density of point defects created by radiation is extremely small and observed effect depends not so much on the dose as on the radiation intensity, it can be assumed that this phenomenon is conditioned by deviation of function of distribution of electrons from thermodynamically equilibrium distribution. The deviation was caused by radiation effect.

Thus, the growth of conduction electrons temperature in metals causes change of interatomic interaction potentials and consequently to reduction of potential barriers for ion movement. In our view the nature of this phenomenon consists in strengthening of screening of ion cores by Fermi electrons.

In paper [9] an attempt was made to calculate the function of electron distribution in nontransition metals under conditions of gamma radiation effect on the basis of kinetic equation and using this function to calculate the metal shear modulus change in pseudopotential approximation. Deviation of the function of electron distribution from that of Fermi was directly taken into account in changing of polarization operator through some component leading to decompensation of direct Coulomb interaction between the ions by means of conduction electrons. Then elastic moduli were calculated based on changes of interatomic interactions nature. The conclusion has been made that change of elastic moduli is not big due to small number of over-Fermi electrons.

We made an attempt to find out the influence of effect of thermal smearing of Fermi surface on potentials of interatomic interaction by way of direct calculations. For this purpose we updated the Lindhard function obtained in the approximation of Fermi sphere, having included the function of conduction electron distribution against pulses into the expression under integral sign [10].

Calculations of interatomic interaction potentials with this function showed that temperature has weak influence on the screening properties of conduction electrons.

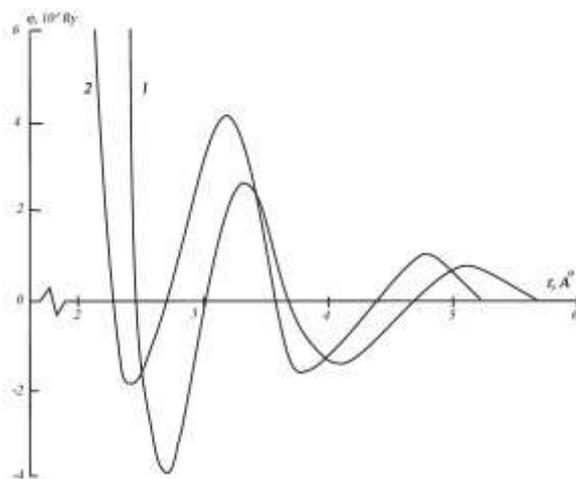


Figure 3. Potentials of interatomic interaction for aluminum: 1- in normal state; 2- the conduction electron pulse exceeds that of Fermi by 10%.

At the same time screening properties strongly depend on concentration of conduction electrons. Figure 3 shows the interatomic interaction potentials for normal state, and for the case, when the valence electrons concentration is 30% higher than the normal one (in this case the electron pulse on

the Fermi surface grows by 10%). Such local growth of concentration can occur in the area of fast charged particle track and lead to “softening” of the crystalline lattice.

3. Conclusion

Thus, the method of calculation of pseudopotential formfactor parameters for additionally ionized ion cores has been developed on the basis of the method of quantum defects. The parameters of Heine-Abarenkov-Animalu formfactor have been calculated and pair potentials of interatomic interaction have been obtained on the basis of these parameters. It was discovered that depth of the first minimum in the potential function of the atom in normal charge state and additionally ionized core decreases. Moreover, for the pair of ionized particles the first minimum in the potential function disappears at all, as a result the particles get on the repulsion branch.

On the basis of pseudopotential approach, the behavior of interatomic interaction potentials under conditions of electronic subsystem excitation has been analyzed. It was discovered that thermal smearing of Fermi surface does not cause any essential change of interatomic interaction forces. At the same time local growth of conduction electron concentration causes “softening” of crystalline lattice when configuration with smaller interatomic distance becomes the equilibrium one. The potentials of interatomic interaction have been calculated for this case.

Thus, the change of interatomic interaction potentials change conditioned by ionized radiation causes different physical phenomena, such as intensification of diffusion processes, sputtering of the surface from the area of fast charged particle track etc. These phenomena can be studied by means of mathematical simulation, particularly, by means of the molecular dynamics method.

4. References

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