Department of Solid State Physics, Lomonosov State University,
Moscow[1]) (a)
and Laboratory of Surface and Interface Physics,
Eötvös University, Budapest[2]) (b)

Electrical Resistivity of Amorphous $Ni_{1-x}P_x$ Alloys Calculated by Molecular Dynamics

By

V.S. STEPANYUK (a), A.A. KATSNELSON (a), O.S. TRUSHIN (a),
P. NAYAK[3]) (a), and A. SZASZ (b)

Introduction   The interest in electrical transport properties of amorphous alloys is caused by a number of their specific features distinguishing them from their crystalline structures. First of all, this relates to the temperature dependence of the electrical resistivity R(T). An anomalous behaviour of R(T) has been found for numerous glassy alloy: temperature increase results in a decrease of the resistivity and this takes place only at definite compositions of alloys. The temperature coefficient of resistivity (α) for glassy alloys with $\rho < 150$ Ωcm has positive value and for alloys with $\rho > 150$ $\mu\Omega$cm α becomes negative /1/. This characteristic behaviour has been found experimentally for $Ni_{1-x}P_x$ alloys as well /2/, with a definite composition where α becomes zero.

Forecasting the electrical properties of amorphous alloys for different component concentrations and different temperatures is possible only in case of having a substantially reliable theoretical access to this problem. Details for fundamental theoretical studies are found in /2, 3/.

In our opinion, one of the main problems in investigations of the electrical properties of amorphous alloys is the determination of the partial structure factors S(q), because the resistivity can be determined by S(q) /3, 4/:

$$\rho = \frac{12\pi \, \Omega_o}{e^2 \hbar \, v_F^2} \int_0^1 d\left(\frac{q}{2k_F}\right) \left(\frac{q}{2k_F}\right)^3 |T_{\vec{k}\vec{k}'}|^2 , \tag{1}$$

$$T_{\vec{k}\vec{k}'} = c_1 |t_1|^2 (1 - c_1 + c_1 S_{11}(q))$$

$$+ c_2 |t_2|^2 (1 - c_2 + c_2 S_{22}(q)) + c_1 c_2 (t_1^* t_2 + t_1 t_2^*)(S_{12}(q) - 1) , \tag{2}$$

---

[1]) Leninskie Gory, SU-119899 Moscow, USSR.

[2]) Muzeum krt. 6-8, H-1088 Budapest, Hungary.

[3]) Working under Indo-USSR cultural exchange programme. Permanent address: Department of Physics, Sambalpur University, Jyoti Vihar, Burla 768019, India.

where $t_i(\vec{k}, \vec{k}')$ is the scattering matrix

$$t_i(\vec{k}, \vec{k}') = - \frac{2\pi\, \hbar^3}{m(2m\, E_F)^{1/2}} \; \frac{1}{\Omega_o} \sum_l (2l + 1) \sin \eta_l^i(E_F) \exp(i\, \eta_l^i) P_l(\cos\theta) \; ; \qquad (3)$$

Ω_o is the atomic volume, v_F is the Fermi velocity, c_1, c_2 are the concentrations of components, S_{ij} are partial structure factors, η_l^i are dispersion phase shifts at the Fermi energy (in the present work, we use η_l^i from /3/). Mostly S_{ij} are determined from the Percus-Yevick equation for the hard sphere model /3/ and the temperature dependence is considered as change in the packing coefficient.

A significantly fast development in molecular dynamics, however, opens new opportunities investigating electronic properties of amorphous systems. A molecular dynamic approach to the Ni-P amorphous system has been successfully applied for structural investigations /5/. Therefore, we apply molecular dynamics calculation for the amorphous $Ni_{1-x}P_x$ system to determine the resistivity in terms of structure factors.

Two compositions have been investigated, the $Ni_{85}P_{15}$ and $N_{75}P_{25}$ alloys. This selection was taken because at phosphorus concentration of $x = 23$ %, the temperature coefficient of resistivity, α, changes its sign /2/. Hence it seemed to be interesting to perform calculations for various temperatures both at higher and lower phosphorus concentrations than the critical one.

The $Ni_{1-x}P_x$ alloy structure has been calculated in the frame of the microcanonic assembly (NVE), where the N (the number of particles), V (the volume of the system), E (the energy of system) remain unchanged. The displacements of Ni and P atoms (total of 256 atoms) in a box was calculated with periodic boundary conditions. The interactions of atoms were modeled by the potentials

$$V(r) = \varepsilon\left[\exp\left(-2A\left(\frac{r}{R_o} - 1\right)\right) - \exp\left(-A\left(\frac{r}{R_o} - 1\right)\right)\right] f\left(\frac{r}{R_o}\right). \qquad (4)$$

The parameters r_o and ε were taken from /6/, and

$$f(y) = \begin{cases} 1 & \text{for } y < 1 \; , \\ 3z^4 - 8z^3 + 6z^2 & \text{for } 1 < y < R_c/R_o \; , \\ 0 & \text{for } y > R_c/R_o \; , \end{cases} \qquad (5)$$

where

$$z = (y - R_c/R_o)(1 - R_c/R_o) \; , \quad A = 3.76 \; , \quad R_c/R_o = 1.4 \; .$$

$)$;   (3)

...ntrations of

...hifts at the

...ly S_{ij} are

.../3/ and the

...nt.

...opens new

...A molecular

...applied for

...calculation

...terms of

...lloys. This

...23 %, the

...seemed to

...higher and

...me of the

...s), V '(the

...nged. The

...culated with

...led by the

(4)

(5)

T a b l e   1

Collected data for the $Ni_{1-x}P_x$ system at T = 300 K

alloy	$\bar{Z}$	k_F (at. units)	E_F (at. units)	ρ_{calc} ($\mu\Omega$cm)	ρ_{exp} /2, 3/ ($\mu\Omega$cm)
$Ni_{85}P_{15}$ Ω_o = 74.91 at. units	1.03	0.739	0.674	108.4 *)	104
	1.03	0.739	0.674	96.1 /3/	
	1.03	0.734	0.674	100.4 /3/	
$Ni_{75}P_{25}$ Ω_o = 74.85 at. units	1.33	0.806	0.778	132.6 *)	175
	1.62	0.862	0.743	160 *)	
	1.33	0.806	0.778	174.5 /3/	
	1.33	0.802	0.771	172.7 /3/	

*) Present work.

Table   2

Calculated resistivities for different temperatures

alloy	k_F (at. units)	T (K)	ρ_{calc} ($\mu\Omega cm$)	sign of α
$Ni_{85}P_{15}$	0.739	1600	117.8	$\alpha > 0$
		300	108.4	
$Ni_{75}P_{25}$	0.862	300	160.0	$\alpha < 0$
		20	167.6	

The integral of motion was solved by the so-called   leap-frog method with 10^{-15} s time step.

In the initial state the atoms were arranged in nodes of an f.c.c. lattice having a Maxwell velocity distribution at the initial temperature of T = 2000 K. Thereafter, during 3000 steps the alloys were thermally stabilized by averaging the thermodynamic and structural characteristics of the system. Starting from this temperature the system was cooled at constant volume in 20000 steps (with average speed of 10^{14} K/s) to reach an amorphous state. The whole process consists of a sequence of cooling, stabilizing, and averaging at some intermediate temperatures, rescaling the atomic velocities after every 10 steps.

At the glass-forming temperature (T_g) - when the solid amorphous state has been reached - the self-diffusion coefficient abruptly decreased.

The radial distribution and partial structure factor $S_{ij}(q)$ were determined for NiP containing 15% and 25% phosphorus. These results were applied for calculations of the electrical resistivity. The essential results are   summarized at 300 K in Table 1.

The valence-electron concentration Z (electrons per atom) as well as k_F and E_F were taken from /3/. The agreement with experiments for $Ni_{85}P_{15}$ is good but for $Ni_{75}P_{25}$ is a little worse. In the frame of the given theory Z could be used as a fitting parameter. An average is taken on the valences of the components /3/, chosen an optimum at $Z_{Ni} = 0.5$, $Z_P = 5$. The recalculated resistivity with this average Z has been evaluated as $\rho = 160$ $\mu\Omega cm$ agreeing well with the experimental results.

Some calculated data for other temperatures are given in Table 2. By increasing the temperature for $Ni_{85}P_{15}$ alloys α is growing but falling for the $Ni_{75}P_{25}$ alloy.

Anomalies in the behaviour of the electrical resistivity can be explained by determining the temperature and concentration dependences of the structure factor by this computer modeling. A simple explanation of anomalies can be given by studies on systematic changes of S(q) /7/.
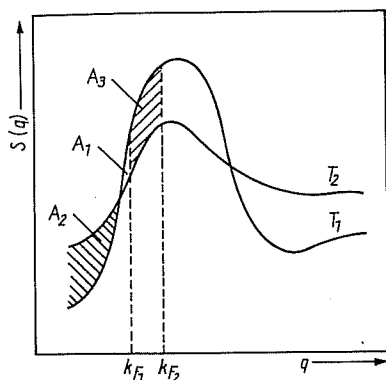
(b) 160

Fig. 1. Schematic change of $S(q)$ of the amorphous alloy for two different temperatures (see text)

The structure factors are schematically shown at two different tempratures in Fig. 1. Increasing the temperature, the maximum of the structure factor decreases, smearing out the shape of $S(q)$, and increasing the half-band-width of the curve.

In the case k_F being in such a position that the area A_2 is greater than area A_1 (k_{F1}) with growing temperature the resistivity will increase as well, $\alpha > 0$. However, at increased phosphorus concentrations k_F is increasing and there could be such a situation when the area A_3 becomes greater than the area A_2 (k_{F2}) (see Fig. 1). In this case the temperature coefficient of the resistivity will be negative.

With this kind of approach it becomes clear why most significant is the position of k_F in relation to the maximum position of the structure factor in similar calculations.

In conclusion, the calculations based on molecular dynamics are capable to describe the resistivity of amorphous alloys as well as to explain microscopically the change of sign of the temperature coefficient of the electrical resistivity.

References

/1/ H. MOOIJ, phys. stat. sol. (a) 17, 521 (1973).
    M. FERNANDEZ, J.M. MARTINEZ-DUART, and J.M. ALBELLA,
    Thin Solid Films 112, L9 (1984).

/2/ J.P. CARINI, S.R. NAGEL, L.K. VARGA, and T. SCHMIDT,
    Phys. Rev. B 27, 7589 (1983).

/3/ L.V. MEISEL and P.J. COTE, Phys. Rev. B 5, 2970 (1977).

/4/ R. EVANS, D.A. GREENWOOD, and P.L. LLOYD,
    Phys. Letters A 35, 57 (1971).

/5/ V.S. STEPANYUK, A. SZASZ, and A.A. KATSNELSON,
    to be published.

/6/ J. LAKKONEN and R.M. NIEMINEN,
    J. non-crystall. Solids 75, 237 (1985).

/7/ L.I. YASTREBOV and A.A. KATSNELSON,
    Foundations of One-Electron Theory of Solids, Mir,
    Moscow 1987 (p. 299).

(Fragments from adjacent column / facing page)

hod with

ce having

2000 K.

aging the

from this

h average

sists of a

peratures,

state has

mined for

lculations

00 K in

k_F and E_F

d but for

used as a

ents /3/,

with this

perimental

increasing

$_5$ alloy.

lained by

ure factor

given by