Electrical and mechanical properties of multi-phase systems under external impacts

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ABSTRACT

The recent progress in creation of materials with negative refractive index inaccessible for natural substances show all-important role of the multi-phase materials in modern technology. In the present work the approach for calculating of effective properties is considered for multi-component composite materials with variable configurations and concentration of inclusions [1]. The approach is based on interpolation formulas obtained between the rigorously calculated limiting borders [2]. The general merit of the model is the ability to obtain algebraic formulas for complicated properties with the vectors of electrical, thermal, magnetic, etc. fields directed along the different axes [3]. The examples of application of the above model are given for the analysis of multi-phase states in the vicinity of pressure-induced phase transition. The model was used for a set of semiconductor compounds like *PbX, SmX (X – Te, Se, S)*, iron ore, etc. [4]. The program for calculation of different electrical, thermal, mechanical etc. properties of *n*-phase systems with variety of configuration and concentration of phase inclusions has been created, which may be applicable for real multi-phase systems.

Keywords: multi-phase systems, effective properties, high pressure, phase transition, variable configuration of inclusion, magnetoresistance, Hall effect, interpolation formulas

1. INTRODUCTION

Multi-phase structures may serve as a model of real layered fabricated devices, the most properties of which being dependent on the concentration and configuration of phase inclusions. The interest to the investigations of these phenomena is connected with proposed application in engineering for the advanced properties, which are inaccessible for uniform materials, for example, low or even negative refractive index [5].

Effective properties of materials (thermal, magnetic, mechanical, etc.) are calculated usually by two main approaches: in the first one the local properties of system are supposed to be known functions of coordinates, and in the second one they are considered statistically as random fields [2].

For two-component isotropic system in the frame of "effective-medium approximation" the equation for "effective conductivity" σ (electrical conductivity, thermal conductivity λ, etc.) was obtained in [6. 7]

$$\sum_{i=1}^{k}\frac{\sigma_i-\sigma}{\sigma_i+2\sigma}c_i=0 \qquad \textbf{(1),}$$

where σ_i and c_i – the conductivity and the concentration of *i*-phase.

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The corresponding equation for two-component isotropic system in the frame of “effective-medium approximation” for thermoelectric power S was obtained in [8, 9]

$$S = \frac{\sum_{i=1}^{k} s_i \sigma_i \gamma_i}{1 - 2\sigma \sum_{i=1}^{k} \gamma_i}, \qquad \gamma_i = \frac{3\lambda c_i}{(2\sigma + \sigma_i)(2\lambda + \lambda_i)}. \tag{2}$$

Fro two-component random inhomogeneous materials the equations for electrical, thermal and galvano-magnetic effects have been obtained also in [10]. The difference of the calculated values of σ and S for the certain binary mixtures by these equations and formulae (1) and (2) was estimated to be several percents [10].

For anisotropic multi-phase systems the effective properties were considered in a lot of works (see, for instance, the references within [11]). The equations for effective electrical resistivity (ρ) of multi-phase systems have been obtained by solving the problem of isotropic dielectric ellipsoid in electric field [12]. In Ref. [2] the expressions were considered for upper and lower bound of effective conductivity of system in cellular model of statistic approach. In the cellular model a total volume is covered by a system of non-overlapping cells in form of ellipsoids. Under chaos packing of ellipsoids a dependence of bounds on form of inclusions is determined by parameters G_i ($^1/_9 \leq G_i \leq ^1/_3$), which are equal to $^1/_9$, $^1/_6$, $^1/_3$ for spherical, needle and disk inclusions, respectively. To take into account a manner of ellipsoids packing, in this approach two additional parameters have been inputted; in some cases similar values for upper and lower bounds of ρ may be received [2]. However, the approach described is very laborious because of enormous awkward calculations. Thus, for various anisotropic systems the different formulas have been derived for calculation of “effective conductivity” (electrical, thermal, mechanical, etc. properties), but the most of them can’t be reducing to the same mathematical form. [11]. So, more suitable and simple models are requested allowing describing the properties of multiphase system with different configuration of inclusions. The application of the above models for the prediction of the outputs of the real electric devices (considered as a kind of multi phase systems) under the external impacts (thermal, electrical, magnetic fields, mechanical stresses) etc. seems to be useful.

2. THE MODEL OF CALCULATION

The calculations have been performed with assistance of a simple but pictorial model of heterophase system (*Fig.1*) [1, 13-14]. Usually, in experiments one can determine only effective characteristics of material (averaged over total volume of substance measured). These effective properties contain “geometrical” parameters of every phase: concentration, shape, and position of inclusions [2, 11, 15].

In our approach an effective electrical resistivity ρ or electrical conductivity (and thermal conductivity λ) are viewed as normalised sums of phase contributions in two equivalent considerations of “consequent” and “parallel” electrical (thermal) connection of phases:

$$\rho = \sum c_i \cdot \rho_i \cdot f_i(\rho) \cdot (\sum c_i \cdot f_i(\rho))^{-1} \tag{3}$$

$$\sigma = \sum c_i \cdot \sigma_i \cdot f_i(\sigma) \cdot (\sum c_i \cdot f_i(\sigma))^{-1} \tag{4}$$

where a sum of phase concentrations c_i is equal to 1 and configuration parameters along electrical (thermal) current are:

$$f_i(\rho) = 3\rho / \left[A\rho + (3 - A)\rho_i\right], \qquad f_i(\sigma) = 3\sigma_i / \left[A\sigma_i + (3 - A)\sigma\right] \tag{5}$$

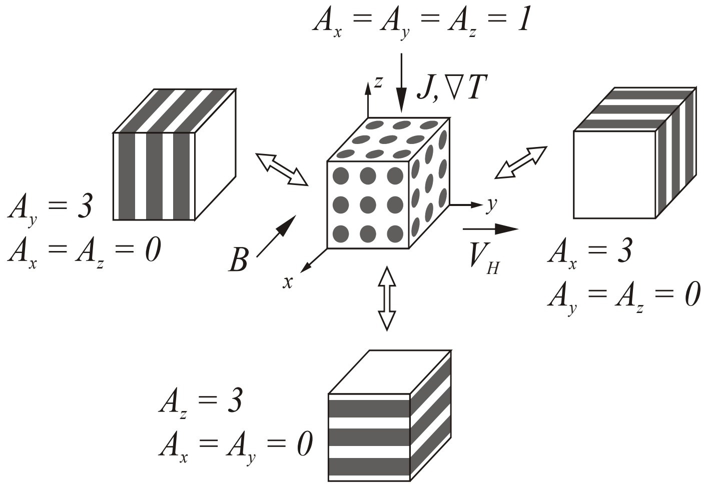

Figure 1. Peculiar cases of materials with various configuration of phase's inclusions (planes). Parameters A_i for different cases are given at the figures. The black arrows show directions of electrical current J (thermal gradient ∇T), magnetic field B, and resulting voltage V_H of transverse Nernst-Ettingshausen and Hall effects. The intermediate shape of inclusions between the limiting cases corresponds to elongated or contracted ellipsoids. The inclusions having the shape of planes may be considered as a stock of needles in the perpendicular directions.

When constant A equals to 0, 3 or 1, the Eqs. (3-4) coincides with cases of parallel, and consequent electrical connections, and with spherical shape of phase's inclusions, respectively (*Fig.1*) [11, 12]. Intermediate values of A ($0<A<3$) correspond to interpolated configuration of inclusions in a certain direction (like elongated or contracted ellipsoids) (*Fig.1*). Unlike the most previous models [6, 11-12] where a shape of inclusions was fixed, in the model a configuration parameter of phase inclusions is variable between extreme limit cases of parallel and consequent (electrical, thermal) connections. The second novel feature of the model is simultaneous consideration and comparison of several properties of material for the same geometrical configuration. Thus, for description of effective properties of phase mixture with arbitrary configuration of inclusions we virtually use well-known formulae for parallel and serial coupling of elements:

$$\sigma_n^{\parallel} = \sum_{i=1}^{n} \sigma_i c_i . \; \sigma_n^{\perp} = \frac{1}{\sum_{i=1}^{n} \frac{\sigma_i}{c_i}} \quad \textbf{(6)}$$

on the assumption of that voltage (in the first case) and current (in the second case) contain additional multiplier G, being a function of concentration and configuration of inclusions: G_i and $G_i(\sigma)$

$$G_i = f_i (\sum c_i f_i)^{-1} \quad \textbf{(7)}$$

$$G_i(\sigma) = f_i(\sigma)(\sum c_i f_i(\sigma))^{-1} \quad \textbf{(8)}$$

Interpolation of formula (2) to the main rigorously calculated limiting borders tends to equation for thermoEMF [1]

$$S = (\sum_i S_i \cdot c_i \cdot f_i \cdot (\rho) \cdot \lambda_i^{-1} \cdot f_i(\lambda)) / (\sum_i c_i \cdot f_i \cdot (\rho) \cdot \lambda_i^{-1} \cdot f_i(\lambda)) \quad \textbf{(9)}$$

And the simple relation follows between the electrical and thermal entities from the above equations (3) and (10) [1]

$$\frac{S-S_2}{S_1-S_2}=\frac{(\rho\lambda-\rho_2\lambda_2)}{(\rho_1\lambda_1-\rho_2\lambda_2)} \tag{10}$$

For isotropic mixture it may be derived from the results of rigorous theoretical calculations in [16, 17].

It's interesting, that Eq.10 allows estimating thermoEMF for any system (including modern nano-structures) without thorough calculations independently on the configuration of inclusions. But the corrected equation for S obtained in the frame of the model is slightly distinguished

$$S=(\sum_i S_i\cdot c_i\cdot f_i\cdot(\rho)\cdot\lambda_i^{-1}\cdot f_i(\lambda))/(\sum_i c_i\cdot f_i\cdot(\rho)\cdot\sum_i c_i\cdot\lambda_i^{-1}\cdot f_i(\lambda)) \tag{11}$$

That tends to deviation from the above relation in the vicinity of the sharp changing of the properties (phase transition point) in case the thermal conductivities of phases sufficiently differ.

As the range of the variation of electrical, thermal, mechanical, etc. properties strongly distinguish, so the thresholds of the transition from the "lower" limit (phase 1) to "higher" one (phase 2) are different for various properties. This tends to visible discrepancy of the data for phase transition recording by different techniques. It may be seen from the relation between S and ρ (Eq. (10) and *Fig.2*). The similar dependences are obtained by using corrected formula (11) for S [3].

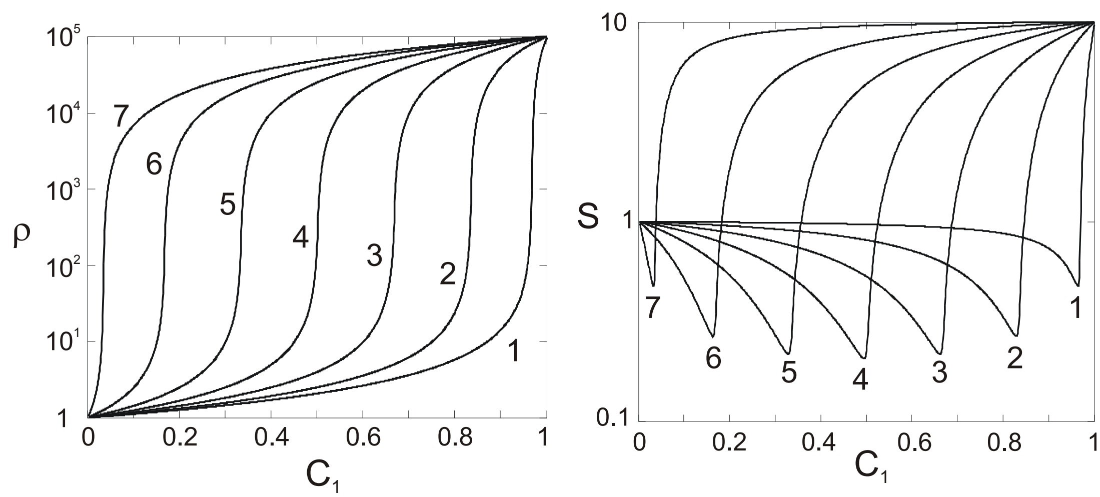

Figure 2. The calculated dependencies of resistivity $\rho(c_1)$ (left plot) and thermopower $S(c_1)$ (right plot) of two-phase system (λ_1=1, λ_2=100, ρ_1=10^5, ρ_2=1, S_1=10, S_2=1 a.u.) on concentration c_1 of phase I. The parameters A are equal to: ***1*** – 0.1; ***2*** – 0.5; ***3*** – 1.0 (spheres); ***4*** – 1.5; ***5*** – 2.0; ***6*** – 2.5; *7* – 2.9.

For two-phase heterosystems Eq. (3) tends to second-order polynomial:

$$A\cdot\rho^2+\rho\cdot\left[\rho_2\cdot\left(3c_1-A\right)+\rho_1\left(3c_2-A\right)\right]-\rho_1\cdot\rho_2\cdot\left(3-A\right)=0 \tag{12}$$

and dependencies of ρ and λ^{-1} on c_i (*Fig.2*) are similar to ones of certain models of spherical and ellipsoidal inclusions [2, 6, 11-12, 15, 18]. For systems of *n*-phases, the equation of n-power on ρ is follows from Eqs. (3)-(4)

3. THE APPLICATION OF THE MODEL FOR VARIOUS TYPES OF IMPACTS

At the real technical multi-phase systems (like IC, microelectronic devices, etc) due to non uniform distribution of electrical and thermal flows as well as the mechanical stresses, the appreciable local electrical and magnetic fields as well as thermal gradients and elastic stresses may arise. Below the influence of these impacts on multi phase systems have been considered using the model proposed and some experimental results obtained at *HgSeS* crystals in the vicinity of pressure-induced "semiconductor-metal" phase transition [19].

3.1 Influence of temperature and magnetic fields on multi-phase systems

The measurements of both the temperature dependence of resistivity and transverse *MR* have been performed in *HgSeS* single crystals on magnetic field *B* up to 2T. The experimental dependencies of *MR* and *R*(*T*) for *HgSeS* initial phases and high pressure phases with cinnabar structure (α -*HgS*) were the initial data for the calculations of two-phase systems properties (Fig. 3-4). The experimental high pressure data for two-phase states of *HgSeS* were comparing with the above calculations (Fig. 3, 4). There is abrupt phase transition *M-SC* at ~1 GPa, when the value of ρ is rising by 6-7 orders of magnitude [20, 21].

By using of Eq. (12) we have calculate $\rho(T)$ and $\rho(\beta)$ dependences; the parameter *A* was varying. The $\rho(T)$ dependences taken for mixture were described respectively by polynomial $\rho(T)=\rho_0+\rho_{01}T+ \rho_{02}T^2$ (*M*-phase) and exponential function $\rho(T)= \rho(0)exp(E_g/kT)$ (*SC*-phase), as the initial semimetal and high pressure semiconductor phases of *HgSeS* and *HgSe* crystals have [21- 28] (Table.1). Here *k* – is Boltzmann constant, and E_g –is activation energy.

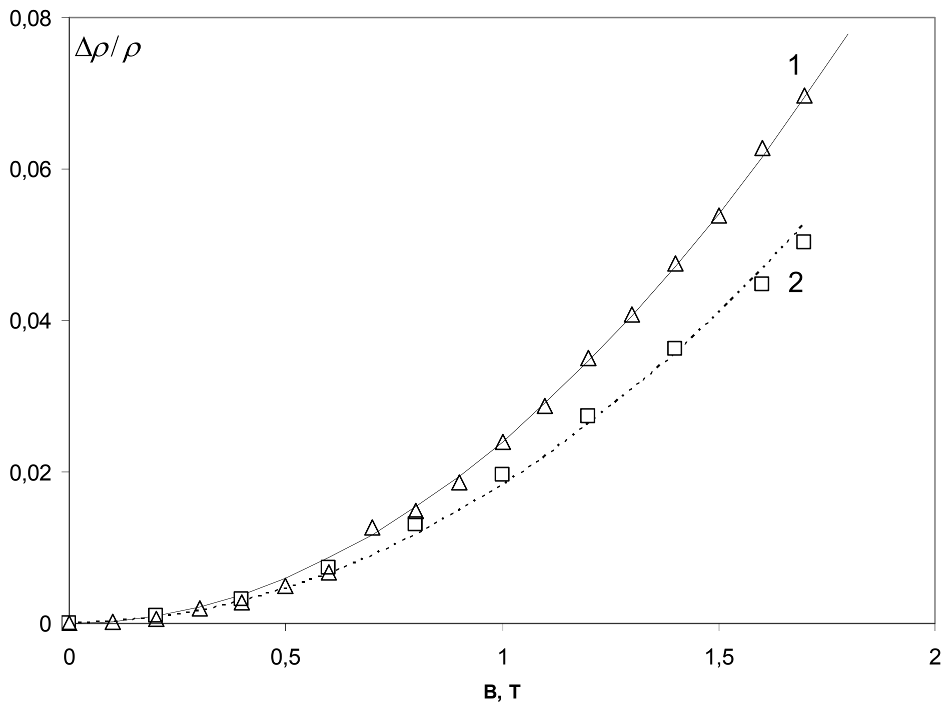

Figure 3.Dependence of magnetoresistance of *HgSeS* crystals on magnetic field *B* at *T*=77 K for *x*=0.104 at *P*=0 (1) and *P*=0.84 GPa (2) (in metastable state [21]); points are the experimental data; the lines are quadratic fitting;the values of ρ for metastable state is higher than at *P*=0 by ~3 orders of magnitude.

Low-pressure (semimetal) phases of *HgSeS* crystals have quadratic positive *MR* [27].

In contrary, high pressure cinnabar phase of *HgSe* and *HgS* crystals have negative *MR* effect. Empirical dependences of negative *MR* were satisfactorily described by the approximate formula $\Delta\rho/\rho = -(zB)^2/(1+(zB)^2)$.

Table 1. Parameters of initial phases and metastable states of *HgSeS* crystals used for fitting of experimental data.

x	*P*, GPa	ρ_0, Ω cm	ρ_{01}, Ω cm/K	ρ_{02}, Ω cm/K^2	*A*	*c*	E_g, eV	*z*, m^2/Vs *T*=293 (77) K
0.104	0	$1.2\cdot10^{-4}$	$-4.3\cdot10^{-7}$	$6.09\cdot10^{-9}$	-	1.0	-	0.13(0.16)
	0.84	$9.0\cdot10^{-4}$	$-4.3\cdot10^{-7}$	$6.09\cdot10^{-9}$	1.0	0.342	0.35	0.09(0.12)

In a weak *B* as positive and negative *MR* according to theoretical predictions [29-32] indeed have nearly parabolic dependence on magnetic field $MR=\pm(zB)^2$, where signs + and – correspond to *M* and *SC* phases, respectively, and parameters *z* (proportional to electrons mobility μ)[29,31] were chosen to be equal to 0,1 and 0,02 m^2/Vs, to fit the experimental values of MR for this phases at B=2T (Fig. 3). So, the most of parameters for calculations were taken from experiment (Table 1). It's interesting to stress that the same set of parameters (A, c, ρ_0) allows to fit the dependences of resistivity both on temperature and magnetic field (Fig. 3, 4). The variation of initial resistivity of phase M (increase of ρ_0) ought to be performed to achieve the accordance with the experiment that corresponds to increasing of density of defects due to phase transformation.

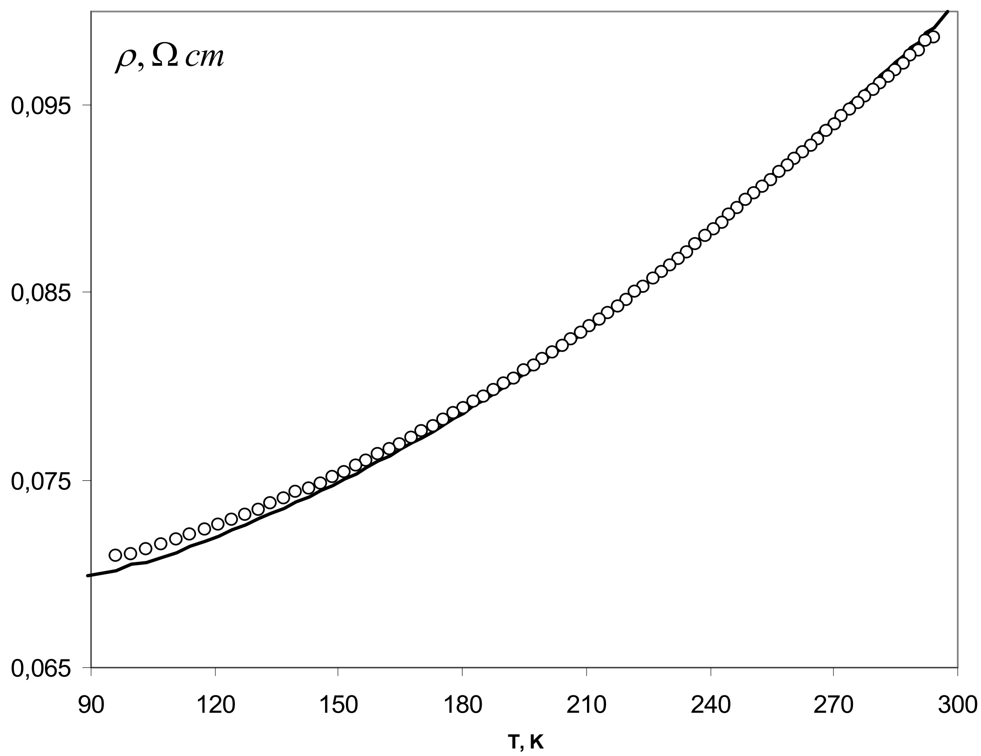

Figure 4. Temperature dependence of resistivity of the same samples *HgSeS* as in Fig. 3 in vicinity of phase transition (metastable states). Symbols – experimental data, curves - calculation by formula (2):x=0.104, P=0.84 GPa, A=1 (Table 1).

3.2 Mechanical properties of multi-phase systems

By using analogy between electrical, elastic etc. phenomena [2] the similar equations may be obtained also for elastic properties. Our formula makes it possible to estimate effective Young's modulus of two-phase composites using their known phase's modulus and volume ratios.

$$E^2(3-A) - E\left[E_1(3c_1 - A) + E_2(3c_2 - A)\right] - AE_1E_2 = 0 \tag{13}$$

For A=0 and A=3 this equation gives well-known formulae describing foliated and fibrous systems [38]:

$$E = c_1E_1 + c_2E_2$$

$$E = E_1E_2/(c_1E_2 + c_2E_1)$$

The dependence of Yung's modulus E on concentration of phases (tungsten carbide) obtained at different papers [33-36] is well fitted by the unit parameter $A\approx1{,}5$ (*Fig.5* and *Fig. 6*), while several different parameters have been used in the papers cited [33-36].

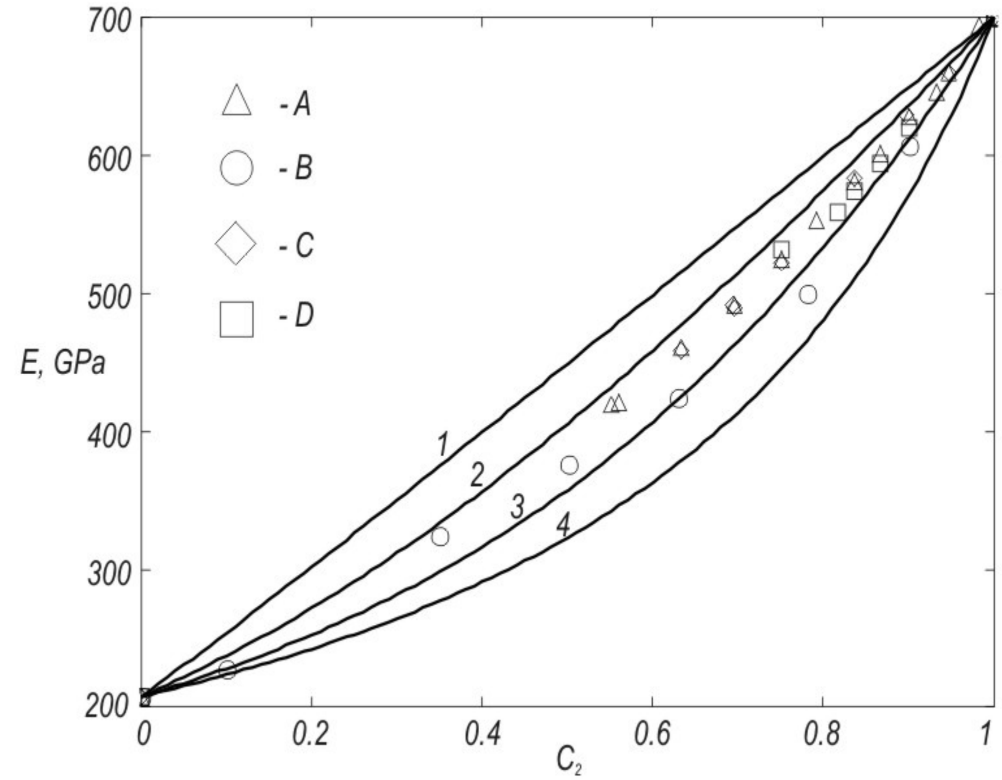

Figure 5. Dependences of Young's modulus of cobalt with particles of tungsten carbide.Curves 1-4 correspond to parameter *A* values: 1 – 0.1, 2 – 1.0, 3 – 2.0, 4 – 2.9. The dots are experimental data from different papers, namely *A* – [33], *B* – [34], *C* – [35], *D* – [36]

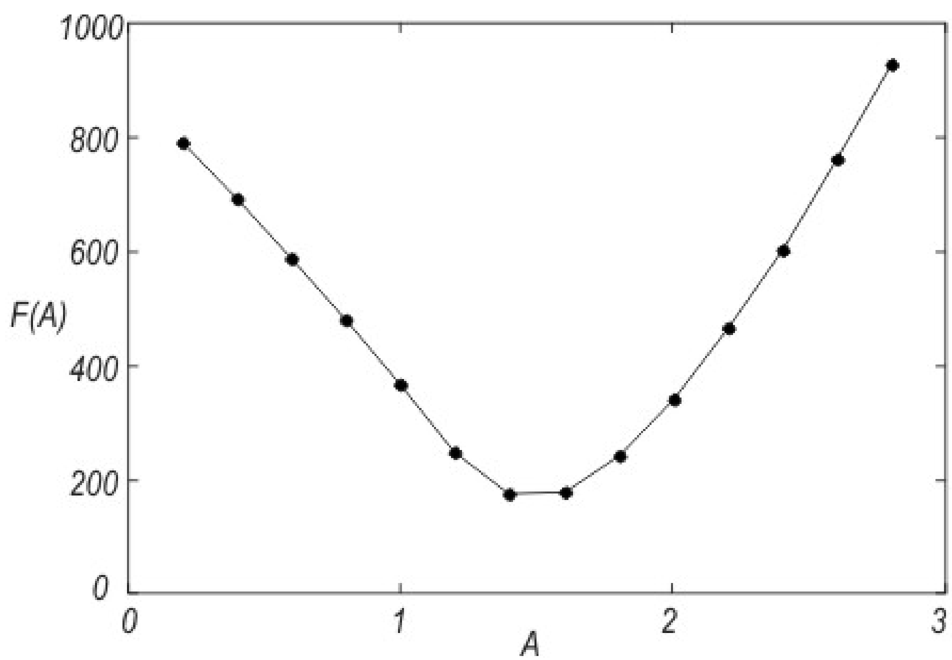

Figure 6.Dependence of function $F(A) = \sum | E_{ex} - E(A) |$, on parameter *A*, where E_{ex} – experimental data, $E(A)$ – calculation results. The best fitting of Fig. 5 corresponds to *A*≈1,5.

3.3 Thermoelectric and thermomagnetic effects at multi-phase systems

By exploring the above approach a coefficient of transverse Nernst-Ettinsghausen (N-E) effect Q and Hall coefficient R for heterophase system may be derived as follows:

$$Q = \frac{\sum_i c_i \cdot Q_i \cdot f_i^T(\lambda) \cdot \lambda_i^{-1} \cdot f_i^H(\rho)}{\left(\sum_i c_i \cdot \lambda_i^{-1} \cdot f_i^T(\lambda)\right) \cdot \left(\sum_i c_i \cdot f_i^H(\rho)\right)}, \qquad R = \frac{\sum_i c_i \cdot R_i \cdot f_i^J(\sigma) \cdot f_i^H(\rho)}{\left(\sum_i c_i \cdot f_i^J(\sigma)\right) \cdot \left(\sum_i c_i \cdot f_i^H(\rho)\right)} \qquad \textbf{(14)},$$

where f_i^T (f_i^J) and f_i^H are configuration parameters of inclusions along thermal gradient ΔT*(current J)* and along Hall direction V_H (perpendicular to magnetic field B), respectively (*Fig.1*).

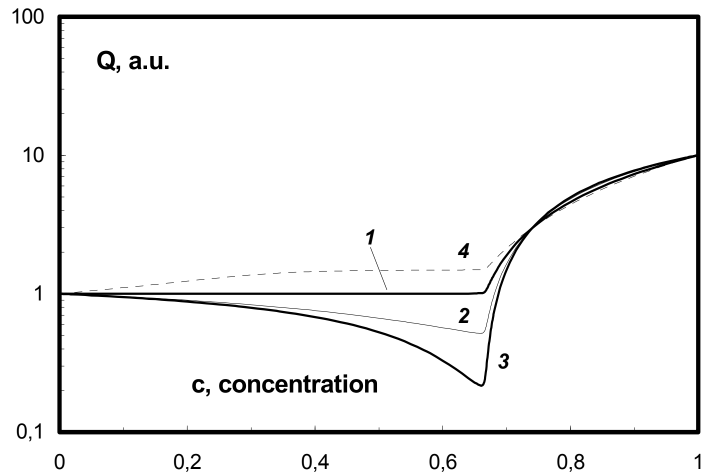

Figure 7**.** The dependencies of transverse Nernst–Ettingshausen effect $Q(c)$ of two-phase heterosystem (ρ_1=10^5, ρ_2=1, S_1=10, S_2=1, Q_1=10, Q_2=1 a. u.) on concentration of phase I in a case when A=1 (spheres). ***1*** – λ_1=1, λ_2=1 a. u.; ***2*** – λ_1=1, λ_2=10 a. u.; ***3*** – λ_1=1, λ_2=100 a. u.; ***4*** – λ_1=1, λ_2=0.01 a. u.

For limit cases of inclusions (see *Fig.1*) Eqs. (14) coincide with the formulae in Ref. [15]. At *Fig.7* the dependence of transverse N-E effect (Q) on composition is shown for various thermal conductivities of phases for a case, when A=1. Using the equations obtained, it is possible to compare behaviour of R and S for real substances (*HgX, PbX, X-Te, Se, Si*) near phase transition points, as well as for layered semiconductor micro-devices [37-38].

Thermoelectric power factor (*æ*) and thermoelectric figure of merit (Z) of heterophase systems:

$$æ = \frac{S^2}{\rho} \qquad\qquad Z = \frac{S^2}{\rho \cdot \lambda} \qquad \textbf{(15)}$$

have to be also functions of phase parameters, as ρ, λ, S depend on concentration and geometrical configuration. One can try to analyse peculiarities of *æ* and Z in two-phase heterosystem. The examples given at *Fig. 8* have shown that thermoelectric power factor of composite is able to exceed significantly values of *æ* of each phase. Loosing a little in the thermoelectric figure of merit on can increase greatly the thermoelectric power factor (*Fig. 8*). Magnetic field can improve additionally the thermoelectric parameters *æ*, Z characterizing the work of thermoelectric cooling devices, as thermomagnetic cooling by N-E effect is known to be preferable in compare with Peltier one. Thus, the approach developed allows to improve working parameters of thermoelectric generators. Thermomagnetic Nernst-Ettinsghausen effect as well as Hall effects may also give rise to parasitic signals at working electric devices.

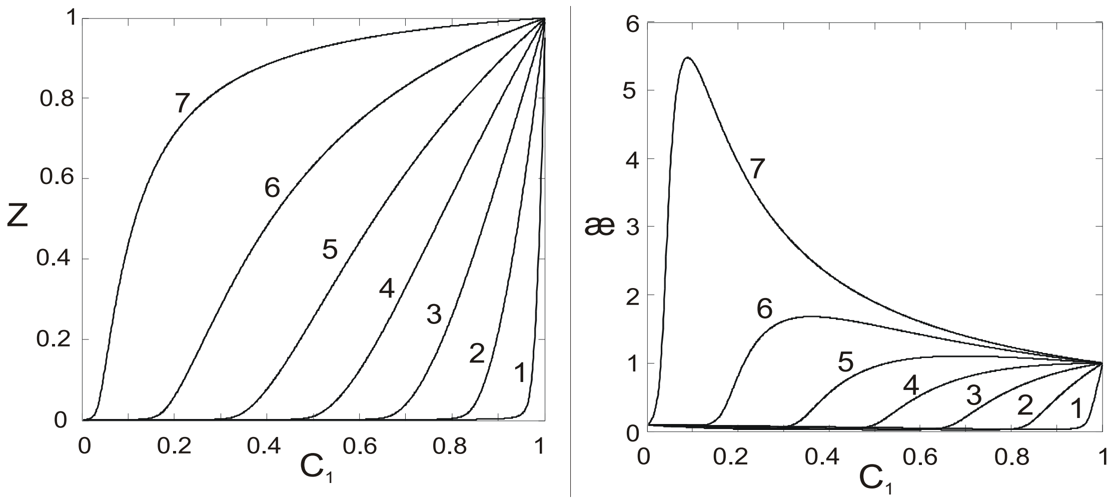

Figure 8. The dependencies of normalized thermoelectric figure of merit (left) and thermoelectric power factor (right) of two-phase heterosystem calculated from Eq. 15 (λ_1=1, λ_2=100, ρ_1=10^4, ρ_2=1, S_1=100, S_2=1 a.u.) on concentration c_1 of phase I. The parameters *A* are equal to: (left): ***1*** – 0.1; **2** – 0.5; ***3*** – 1.0 (spheres); ***4*** – 1.5; **5** – 2.0; ***6*** – 2.5; **7** – 2.9; (right): ***1*** – 0.1; ***2*** – 0.5; ***3*** – 1.0 (spheres); ***4*** – 1.5; ***5*** – 2.0; ***6*** – 2.5; *7* – 2.9.

4. CONCLUSION

The traceable pictures demonstrating the behavior of some of considered properties of two-phase systems as a function of concentration and configuration are shown at Fig. 9 and Fig. 10

The mathematical equations for the thermoelectric power, resistivity, Hall coefficient, and for thermomagnetic and elastic effects, obtained in framework of the model developed explain visible discrepancies of experimental data of high-pressure investigations of materials. In particular, shifts of phase transition pressure obtained by using of various structural, electrical, thermal, magnetic and combined effects may be clearly described. The model may be applicable to the real object (like IC, electric devices) presenting multi-phase systems with the certain configurations.

The main two features of the approach are: the wide variability of the configuration (shape of inclusions) as well as their concentrations, and the consideration of several properties of multi-component material simultaneously in the frame of the same mathematical model. The general merit of the model is the ability to obtain algebraic formulae for complicated properties with the vectors of electrical, thermal, magnetic, etc. forces directed along the different axes.

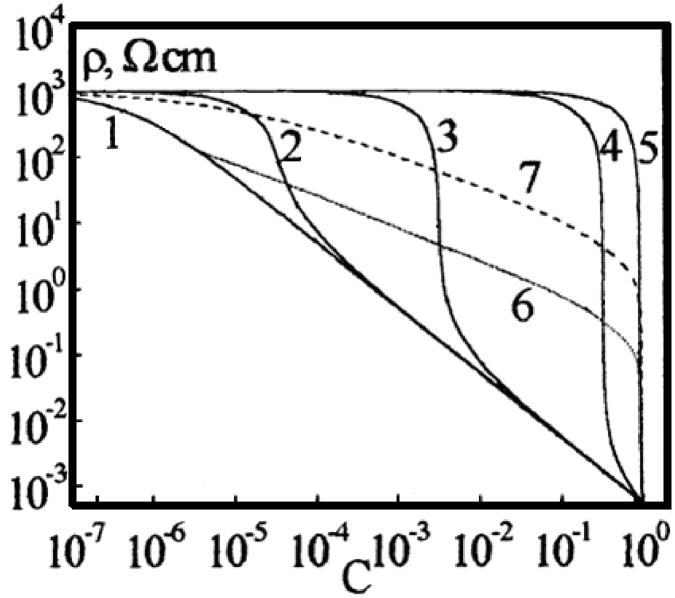

Figure 9. Dependences of resistivity of two-phase material (solid curves l-5) and boundaries of sign inversion (dash curves) of temperature coefficient *ρ* at *T*=300 K (6) and *MR* at *B*=2 T (7) on *M* phase concentration *c* for various configuration parameters of inclusions *A*:1 -A=l0^{-16} (parallel connection of phases, "needles"); 2 - A=10^{-4} ; 3 - A=10^{-2} 4 - *A*=l (spherical inclusions); 5 - *A*=3 (consequent connection of phases, "layers"). Parameters of *M* and *SC* phases correspond to data for *HgSeS* (*x*=0.104) sample and *HgSe* high pressure phase [20, 28]; the values of *z* parameter are equal for *M* and *SC* phase respectively: 7 - 0.1 and 0.02. The region between curves 1 and 6 corresponds to states possessing metal type of conductivity.

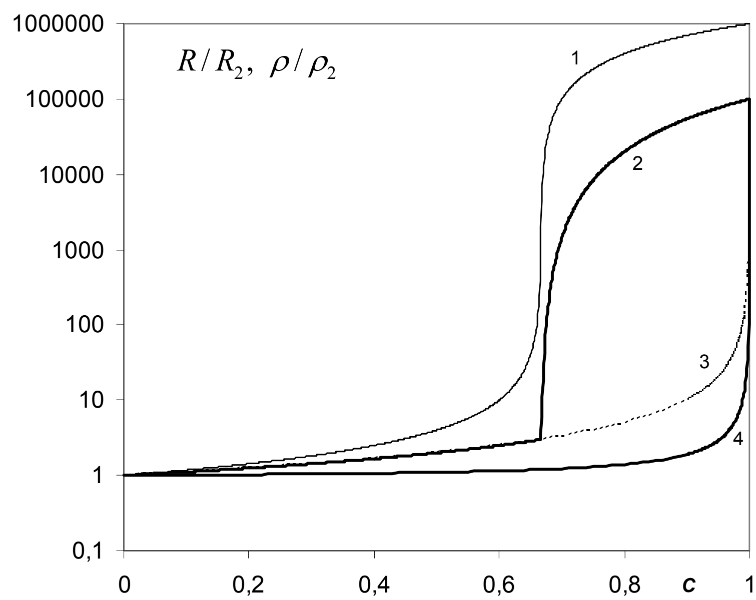

Figure 10. Calculated dependencies of normalized values of resistivity (curve 1) and Hall constant (curves 2-4) on concentration of semiconductor phase *c* for some peculiar limiting cases of inclusion configuration: 1, 2 – spherical inclusions ($A^J = A^H$=l); 3 - layers, oriented along the *J* and Hall direction (A^J=A^H=0); 4 - layers, perendicu1ar to *J*-(A^J=3, A^H=0), or Hall directions (A^J=0, A^H=3).

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