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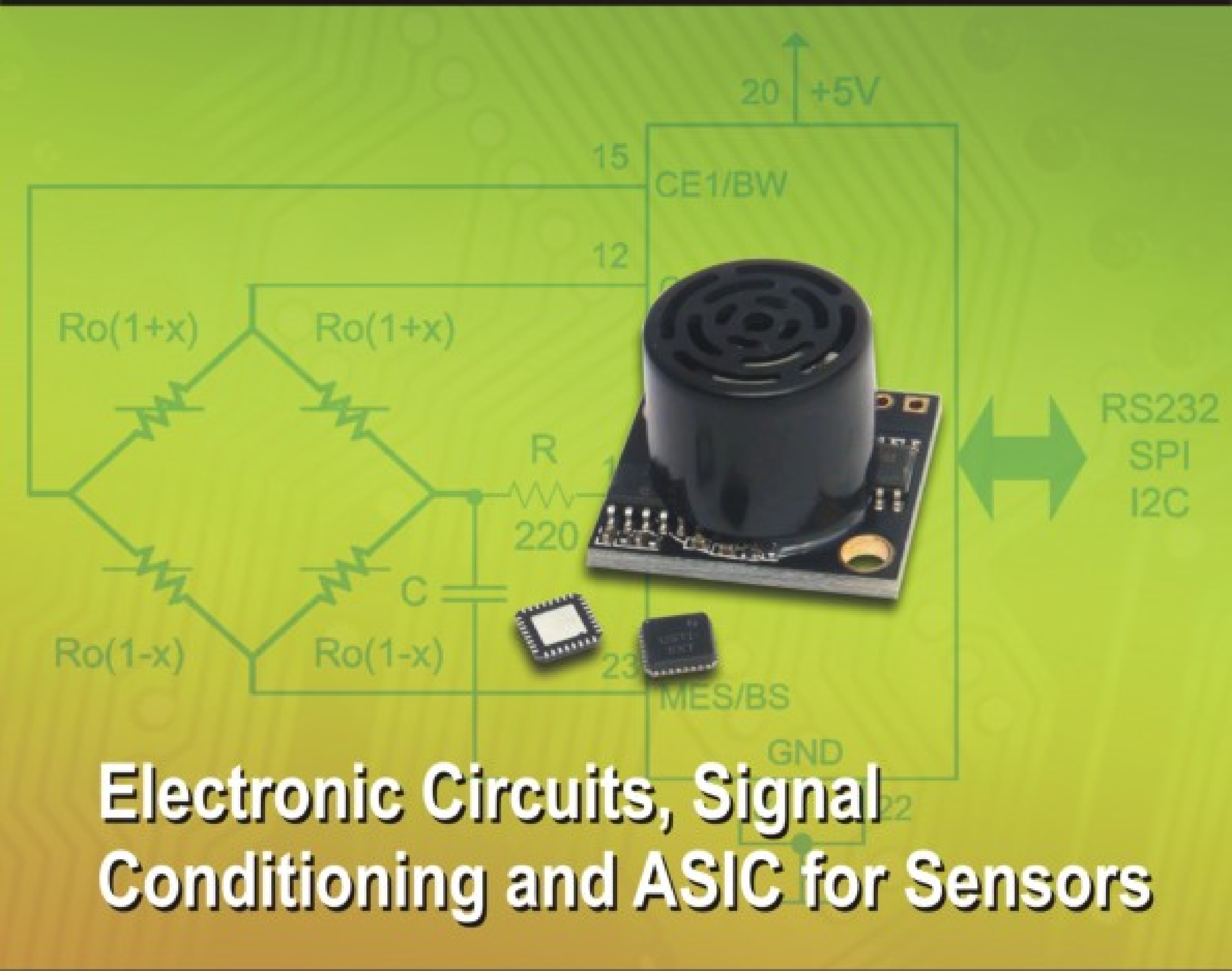

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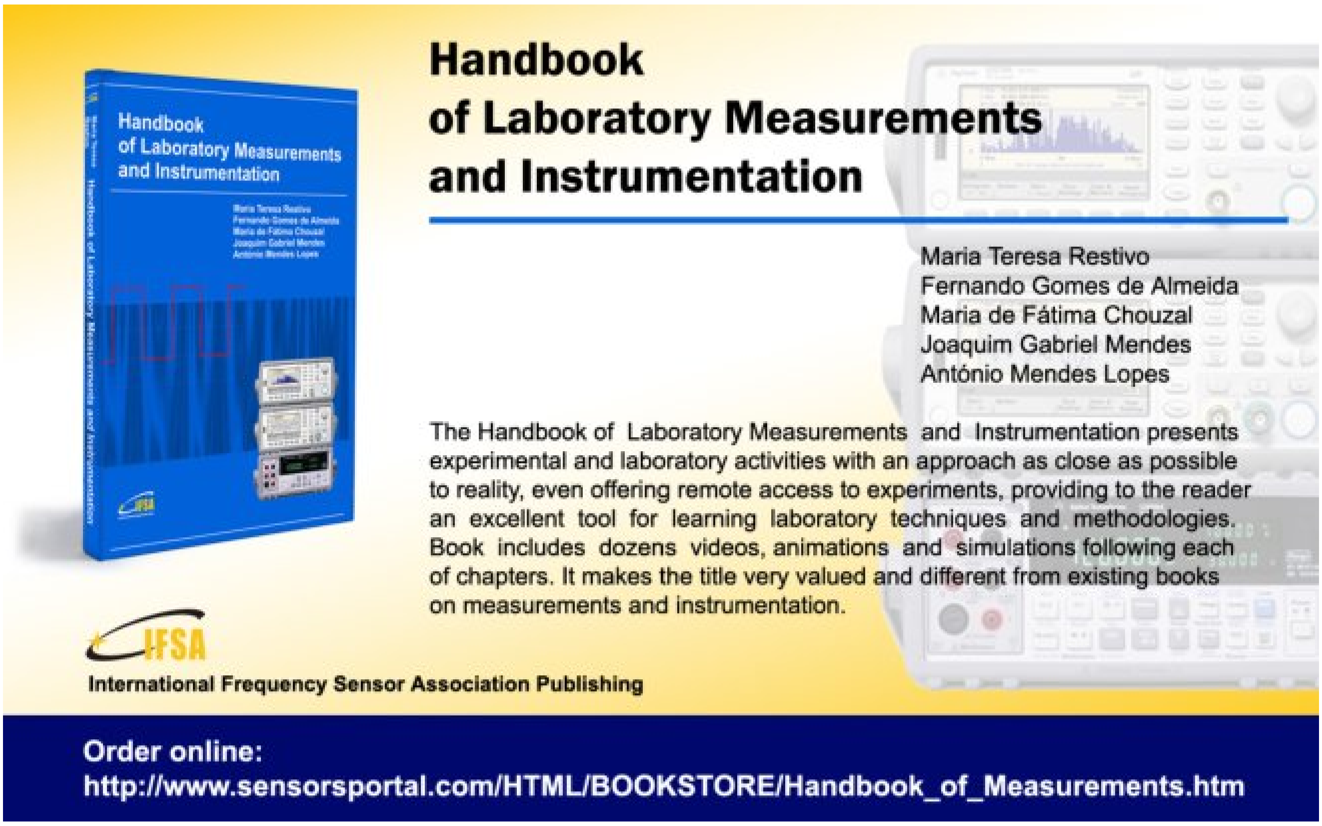

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Research in Nanothermometry.
Part 4. Amorphous Alloys of Thermo-resistive Thermometry

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Abstract: Development of thermometry demands the continuous to materials research. It is the main factor ensuring the high reproducibility of nanotechnology research. Therefore, the investigations of sensitive elements of resistance thermometers are becoming quite important. Amorphous alloys have been studied and the results discussed on a model level. The nanothermodynamics is suggested to be efficient in the analysis of metallic glass structural peculiarities and electrical properties. *Copyright © 2012 IFSA.*

Keywords: Resistance thermometer, Sensitive element, Metallic glass, Nanostructured material.

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1. Introduction and Task Definition

With the advancement of techniques of creating and characterizing materials, a huge amount of ‘‘small’’ size grains (micrometers), nanosystems, molecular magnets, and atomic clusters, have been formed and displays a variety of interesting physical and chemical properties. It is important and timely to develop the new theoretical tools, to address experimental findings. On the other hand, the rapid progress in the synthesis and processing of materials with the structures at the nanometer size has created a demand for greater scientific understanding of the thermodynamics on nanoscale [1]. Nanostructured materials including metal alloys with amorphous structure, for instance, metallic glasses (MG) could be also regarded in this manner. The new-gained materials with the high specific resistance ρ at the small value of its temperature coefficient α foster their study.

The particular efforts are made in the endeavour to bind the electrical MG properties with the peculiarities of their manufacturing technology, which could be profitable in gaining the special electrotechnical materials [2], spintronics [3], etc. Moreover, very small particles have special atomic structures with discrete electronic states, which give rise to certain properties in addition to the super-paramagnetism behavior [1]. The study of nanostructured materials reveals the fact that the unique physical properties could be gained not only by the constitution change but the varying of warranty conditions and consequent thermoprocessing that affects the structure to a great extent. [4].

2. Object of the Work

The research of electrical properties of nanostructured materials (for example, metallic glasses $Fe_{40}Ni_{38}P_{13}B_9$ in terms of the peculiarities of manufacturing technology evaluated within the phenomenological nanothermodynamics).

3. Subject of the Research

Tetraric metal amorphous alloys consisted of two transitive ferromagnetic metals (*Fe*, *Ni*) and two amorphizators (*B*, *P*) form the content of $Fe_{40}Ni_{38}P_{13}B_9$. MG $Fe_{40}Ni_{40}P_{14}B_6$ similar in composition has been studied in details [5].

4. Experimental Part

In general, five groups of patterns are gained as MG that at the moment of casting differed by (a) the alloy temperature T_p, (b) the rotation speed of the massive disk covered by the alloy at the moment of casting *n*, and accordingly by (c) the thickness of the gained foil δ (Table 1).

Table 1. Main technological MG parameters and thickness.

Pattern group number	T_p, K	T_{cr}, K	$n, 1/\min$	$\delta, \mu m$
1	1373	653	1000	100
2	1373	653	3000	30
3	1273	653	3000	30
4	1523	653	3000	30
5	1100	653	2000	60

Two groups of experimental dependencies *R*(*T*) and α(*T*) (Fig. 1) were formed (Fig. 1). Measurement was conducted according to the known methods in the reference points: 4.2 K, 13.8 K, 20.4 K, 77.4 K and 300 K. The error of electric resistance measuring was made ±0.005%, i.e. the reliability of the first four signs of the measured value was assured. The processing of the gained result was made with the help of the Curvescript software.

Experimentally gained dependencies *R*(*T*) are approximated (error $\leq 10^{-3}$% at the correlation factor 0.99999998) by polynomials of the 3rd degree: $R(T) = A + BT + CT^2 + DT^3$. To compare, the description by the polynomial of the 2nd degree has given the error 1.6% at the correlation factor 0.998.

The values of factors *A, B, C* and *D* for the researched MG groups ($R1(T) - R5(T)$) are given in the following equations (1).

$$\begin{aligned}
R1(T) &:= 1.9426 - 0.0002635 \cdot T + 4.62 \cdot 10^{-6} \cdot T^2 - 9.59 \cdot 10^{-9} \cdot T^3 \\
R2(T) &:= 1.7518 - 0.0002268 \cdot T + 3.98 \cdot 10^{-6} \cdot T^2 - 8.26 \cdot 10^{-9} \cdot T^3 \\
R3(T) &:= 1.3109 - 0.00012 \cdot T + 1.84 \cdot 10^{-6} \cdot T^2 - 3.07 \cdot 10^{-9} \cdot T^3 \\
R4(T) &:= .958 - 0.00012 \cdot T + 2.22 \cdot 10^{-6} \cdot T^2 - 4.35 \cdot 10^{-9} \cdot T^3 \\
R5(T) &:= .995 - 0.0000642 \cdot T + .844 \cdot 10^{-6} \cdot T^2 - 1.11 \cdot 10^{-9} \cdot T^3
\end{aligned} \tag{1}$$

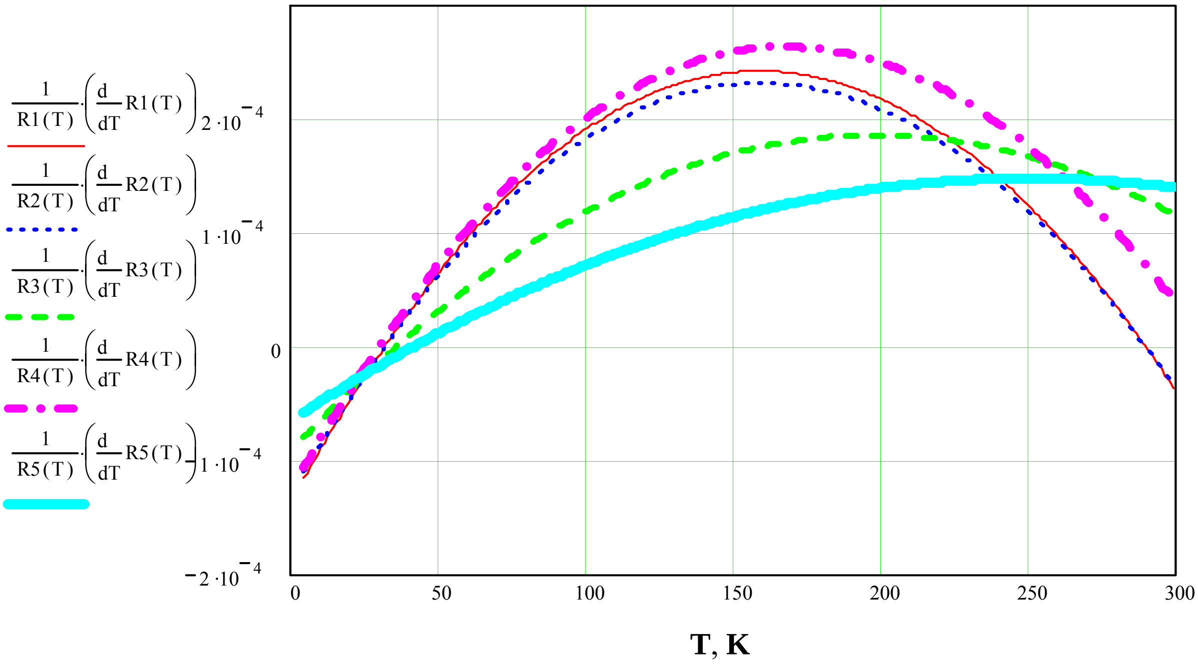

Fig. 1. Temperature dependence of the temperature factor of electric resistance α of the researched metal amorphous alloys.

The character of the temperature dependencies $R(T)$ and $\alpha(T)$ affirms the following fact: the remaining MG resistance at the low temperatures is considerably larger as compared to their crystalline analogs; the temperature coefficient of the MG resistance – on contrary, less, and could be $\alpha > 0$, $\alpha < 0$ and $\alpha = 0$; low-temperature minimums pertain to temperature dependencies of MG electric resistance, and maximums at 160 ... 260 K (Fig. 1) pertain to α.

5. Model-theoretical Argumentation

According to [1, p.106], the described above character of nanostructured material manufacturing corresponds to the MPNUR-mechanism of forming the metastable structural state of a M-phase as well as organizing the new B-region of a pseudostable M-phase consequently of the additional substance-nanostructuring pressure effect. The existence of distension areas in MG with considerably smaller material density – defects of the n-type as 12-20 atom-sized clusters with material density, approximately 10 % lower as compared to the unified characteristics – is affirmed by our investigations [6]. Their number as well as the number of symmetrical contraction defects could be estimated at the level of 2.5 % from the general volume. During 30 minute annealing at 623 K, 10 % of defects recombine. Moreover, the inner mechanical strains reach 15 MPa and decrease sevenfold while annealing. Density fluctuations caused by the nonuniform distribution of free volume in a pattern could become the sources of inner strains.

Structural relaxation attributed to the changes caused by the reformation of a local atom structure is researched in amorphous and crystalline alloys $Fe_{40}Ni_{40}P_{14}B_6$ at the temperature from 340 K (start) to 600 K (end) [7]. It is shown that this phenomenon is related to the disappearance of free volume in an amorphous structure. The study following the method of a quadruple fission has shown that a local redistribution is made around the atoms of *Fe*, consequently of what the temperature of Curie rises considerably in ferromagnetic alloys on the basis of iron ($Fe_{40}Ni_{40}P_{14}B_6$).

The results of relaxation processes study at the low temperatures 1…30 K, where the changes in sound speed are referred to the electronic states of a two-level system are of interest. The groups of atoms are remaining in two states: in one potential hole or in another, isolated from the former by a potential barrier. The difference of energies between those states is 2ε, moreover $\varepsilon^2 = \Delta_0^2 + \Delta^2$, where Δ_0 is the energy of an overhead cover, 2Δ is the asymmetry energy. The energy of an overhead cover is $\Delta_0 = h\omega e^{-\lambda}$, where $\lambda = \left(2mv_0/h^2\right)^{1/2} d$; ω is the fluctuation frequency of an atom of the mass *m* in the potential minimum; d is the width of system.

Two-level systems cause the considerable influence on the spreading of phonons. Phonons of the deformation field of a resilient wave modulate the energy *ε*, and in case $h\omega \approx \varepsilon$, the phenomenon of absorption appears in the system. The excited state arisen could relax either while excitement intensifying or spontaneous transferring. Therefore anomalies of sound speed in MG could be explained by resonance / relax absorption in a two-level system. The contribution of a relax process is getting less at temperature fall which leads to the increase in sound speed and to other similar anomalies, e. g., to the minimum of inner friction, etc.

The effects like these confirm the existence of fields of considerable mechanical microtensions whose influence is equal to the doping by a number of admixtures. In this case, we should apply nanothermodynamics [1] in order to explain the MG electrokinetic and other properties. The latter stipulates the introduction of two additional freedom degrees into the main equation of thermodynamics: σdM (*M*- the surface area), caused by the superficial tension σ, and γdV, caused by the expenses of the specific energy γ for the formation of precipitations of the second phase with the volume *V* in the matrix of output substance.

5.1. Consideration of Experiment Results

First of all, let us ascertain the reason for the description of the researched temperature dependences of electric resistance by the four-member polynomial of the third order. All other standard functions of 16 types, involved into the ***Curvescript*** software, have proved to be incapable of describing the gained results in a proper way.

A. Let us consider the two-phase MG model, i.e., assume that there are precipitations of other phases in a matrix, e.g. pseudo-phases which could be represented by the mentioned above microvolumes with different density. If the specific electrical resistance of a matrix is described by the equation $\rho_1 = a + bT$, according to the rule of Mattison, and that of another similar phase is represented by the equation $\rho_2 = c + dT = a + bT + a' + b'T = \rho_1 + \Delta\rho$, and taking into account that the volume part of the second phase makes $\Delta V/V$ on the efficient intersection area $q = \Delta S/S$, we gain the equation of the third order for the specific electrical resistance of two-phase material:

$$\rho_a(\Delta\rho)^2 = (1-q)\rho_1(\Delta\rho)^2 + (\Delta\rho)^3 - q\rho_1^2, \tag{2}$$

Conducting the analysis of factors of its components, we come to the expressions:

$$A = a'\left[(1-q)aa' + (a')^2 - qa^2\right],\ D = (1-q)b(b')^2 + (b')^3 - qb^2b' \tag{3}$$

At the $q = \Delta S/S$ being equal to 0.1, the factor A is larger than 0 at $a' \geq 0{,}1a$ that meets realities. Simultaneously the factor D, experimentally determined to be lower than 0, is negative in the case of slight deviations from the linearity of electric resistance temperature-dependence $b' \leq 0{,}1$. The comparison of the factors B and C allows to represent that if $B < 0$ then $C > 0$, and the factor A is proportional to the factor C having the same sign; the factors B and D stay in the analogical dependence.

The following issues are: the basis of MG is homogeneous material manufactured due to the usual technologies (avoiding its quenching and the following thermoprocessing); $\rho = a + bT$, where the factors *a* and *b* do not depend on the manufacturing technology. Whereas the factors $a';b'$ as well as *q,* characterizing the precipitations of real MG, depend on the manufacturing technology. The increase of the constant q corresponds to the decrease of the factor A – the first member in the temperature dependence of electrical resistance. It is supported by the experimental data where A decreases from 1.9426 through 1.7518 till 1.3109 at the increment in the temperature of fusion before pouring out on the disk from 1273 K through 1373 K till 1623 K.

Explanation is that the specific volume depends on the speed of chilling, determined due to the mentioned temperature. The increment in the MG volume reaching some percent and considerably influencing the transmission processes, occurs with temperature rising. Moreover, the structures with less average density have higher values of speed transference. The first member of the expression (1) behaves itself in the same way at the decreasing of rotation speed of the disk where quenching is taking place. So at 3000 rpm (thickness 30 μm), it makes 1.7518; at 2000 rpm (60 μm) - 0.995, and at 1000 rpm (100 μm) - 0.958.

B. Let us compare our investigations with the works made in the Saint Petersburg's school of deformation physics where the decrease in the real limit of material strength concerning the theoretical values is explained by the presence of tensile defects [8] as micro- or nano- concentrators of mechanical tensions. According to the results of analyzing the spectrum of combinational light dispersion 632 cm^{-1}, the appearance of dot radiators with the temperature in 2...5 times prevailing the average is fixed in the transparent polyethylene terephthalate in the presence of extention deformations or without them. The availability of low- and high-frequency satellites with reference to the main maximum (see Fig. 2 with maximums at 624 and 640 cm^{-1} respectively) confirms the existence of micro- and nanoareas of extension and contraction, respectively. The displacement of frequencies for 8 cm^{-1} is explained by the deformation of interatomic bonds of tensile quasi defects for the value $\varepsilon = -\frac{1}{G}\frac{\Delta\nu}{\nu}$**,** where G is the mode parameter of Gruneisen**,** $\Delta\nu$ is the alteration of light frequency.

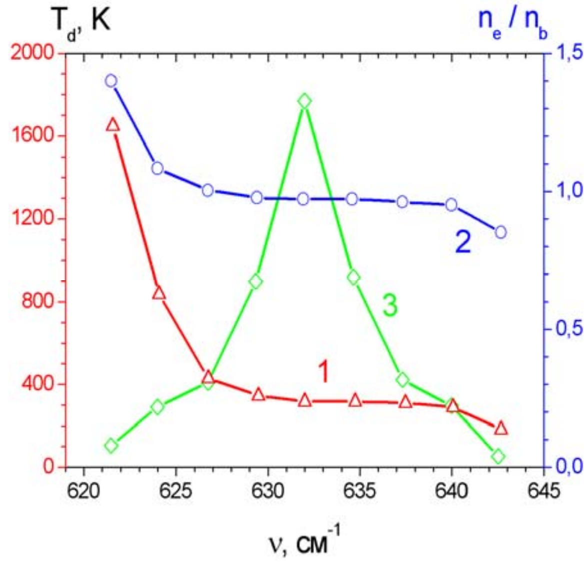

Fig. 2. Dependence of local temperature (1) and phonon occupation value (2) vs. the deformation of interatomic bonds in polyethylene terephthalate (3) - the shape of relative intensity of Raman spectrum.

While capturing the phonons by the tensile quasi defect, its local temperature is rising, considerable thermo-extension takes place that in some moment could cause the rapture of interatomic bonds and thus microcrack appearance. Using the intensity of fluctuation stripes in Raman spectrum of combinational dispersion (Stokes and Anti-Stokes bands), the phonon value or capture by tensile quasi defects were determined. If usually they are much less than equilibrium, determined from the formula of Bose–Einstein, then in the case of the extension effect they are on contrary much higher. Local temperature of the above mentioned quasi defects (with linear size till 10 atoms) is determined as $T_d = \frac{h\nu}{k\left[\ln(n+1) - \ln n\right]}$. Regarding to the dependence given in the Fig. 2, it is obvious how essential the dot temperature increasing is on these defects, being especially important for the thermosensitive substance.

Micro concentrators serve as the trigger that launches the transformation of the energy applied to the material into microcracks. The appearance of the latter is indissolubly related to the emission and redistribution of energy, considerably exceeding the energy capacitance of elementary deformation acts. The indication of tensile quasi defects as physically elementary nanosized subsystems of structure enables us to stipulate the introduction of thermodynamic values i.e. the application of nanothermodynamically nonequilibrium processes to the researched substance.

Considering the mentioned defect in its closest surroundings as locked thermodynamic system, we could show that the production of entropy within this system is determined by the sum of products of correspondent thermodynamical powers and fluxes. The product of tensor gradient of mechanical tension $\nabla\bar{\sigma}$ and deformation tensor $\bar{\varepsilon}$ could be regarded as the main among them. As a result, the electrotransference parameters change, leading to the increment in electric resistance as compared to the resistance of a crystal pattern, according to [9].

The reasons to treat the given substance as metal glass disappear at the removing of mechanical tension gradients nearby the defect, at least because of electrotransference. The immediate crystallization is taking place, moreover, the proceeding to this process is described by the diffusion factors: the temperature exposure T_{exp} and the time $t_{p.cryst}$ of maturing at this temperature without crystallization:

$$t^2_{p.cryst}T_{\exp} = 600...800Bt_c\,, \tag{4}$$

where t_c is the crystallization time, studied in the process of researching its dynamics; B is the introduced criterion called the temperature exposition: $B = 0,75T_g t_{an}$; T_g is the glass-forming temperature; t_{an} is the temperature of isothermal annealing.

6. Conclusions

1. Precision analysis of temperature dependences of electric resistance of $Fe_{40}Ni_{38}P_{13}B_9$ metallic glass enables us to evaluate their structure as structure of heterogeneous two-phase materials (nanostructured pseudo-selection in a homogeneous matrix) that is the issue of quenching in the manufacturing process.

2. The researched materials differ by the null value of an electric resistance temperature factor, and therefore could be employed for manufacturing the primary precision resistance thermometers and strain gauges, whose deformations influence is increasing with the fall in the electroresistance temperature factor.

3. Nanothermodynamics is suggested to be efficient in the analysis of metallic glass structural peculiarities and electrical properties.

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