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Sergey Y. Yurish



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Topic E2: Transportation & Mobility

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Research in Nanothermometry. Part 8. Summary

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Abstract: Recent advances in nanotechnology are expressed by the atom-scale insights successes related to development in nanometrology. Its main integral part is supposed to be nanothermometry. The latter rests on nanothermodynamics while determining its advancement as well as the overall progress of nanotechnology. This mutual feedback, described by the examples of certain types of thermotransducers in a series of articles published in *Sensors & Transducers* journal earlier, is under consideration in this paper. *Copyright © 2012 IFSA.*

Keywords: Nanothermometry, Nanometrology, Nanothermodynamics, Fluctuations, Noise

1. Introduction

Succession of investigations in reference to the study of processes in thermodynamic materials for the purpose of creating the high-precision thermometers adapted for extreme temperatures and other exploitation conditions has been made through the last years [1]. The required experience, encouraged by the endeavor to cognize the nature of measuring instrument drifting which is related to the processes in a thermometric substance, proved to be precious, since being concerned with the profound processes in the substance, namely at micro- and nano-levels. Besides, with the development in nanotechnology, nanometrology [2] accompanied by nanothermodynamics has arisen [3].

Metrology is inseparably related to thermometry. The same concerns nanothermometry as an integral part of nanometrology. Its development implies both a direct task, to measure temperatures and other substance thermo-properties in nanoworld, and an opposite one, to ascertain the reasons for particular

behavior of metrological characteristics, linking them with fluctuation-dissipation processes in thermometric substance during manufacturing and exploiting at a micro- and nano-level, on the basis of high experimental experience and metrological culture due to the measurement results with the use of nanosized and nanostructured sensitive elements of thermometers.

In nanotechnology the term "Temperature" acquires ambiguous statistical thermodynamic sense [4]. Hereby, the absence of knowledge about the temperature of the researched micro- and nano-objects does not allow to assure the reproducibility of technologies, and measurement leads simultaneously to the shift of a temperature field, appearance of methodical error of a measurement instrument and sometimes to the destruction of a nanopattern despite of applying contact and/or remote method [5].

Keeping the deep insights of metrological approaches into the essence of processes within thermosensitive substance, and mastering the achievements of nanotechnology, we have widened the spectrum of thermometric methods: assimilating Raman thermometry [6-7] of micro- and nanopatterns; conducting the research in thermometry based on solid- and liquid-phase sensitive elements at decreasing their sizes down to a nanoarea; and studying the role of a thermosensitive substance superficial tension gradient as a main thermodynamic force of nanothermodynamics in the forming of metrological characteristics [8] besides the already ascertained role of a mechanical microtension gradient [9] in the forming of thermometer transformation function drift.

The branches of thermometry were singled out in order to study the nanostructured thermosensitive substances for the purpose of improving durability or increasing the temperature limits of sensors. Those are the branches of ultrasonic thermometry [10], noise thermometry [11] and resistive thermometry [12]. Owing to the means of engineering maintenance including the traditional equipment for calibrating, thoroughly conducted resource tests, and row of structural methods for characterizing a structure and its changes during exploitation, we managed to associate the effect of nanostructuring with quite precisely controlled micro- and macro-characteristics.

The widest investigations were made in traditional thermoelectric thermometry [13], where thermodynamics of irreversible processes, having proved mutual independency of the chosen thermodynamic coordinates (forces and flows) with regard to physical phenomena spectrum, is combined with metrology (its integral part - thermometry) and the relevant apparatus of "correlativeness / incorrelativeness" of the gained measurement results with the row of influence factors.

All the papers on the subject are published in the series of articles including Research in Nanothermometry, Part 1-7 [4, 5, 7, 8, 10-12] and [6, 9, 13, 14] being completed by this work where theoretical aspects and possibilities of the further progress in nanotechnology that follow from the conducted research are considered.

2. Task Definition

The fundamental issues in this domain of nanomaterials are [3]: ... ability to obtain the required composition, not just the average composition but details such as defects, concentration gradients, etc., and to control the modulation dimensionality … More specifically the following issues have to be considered for the future development of nanomaterials: … better understanding of the influence of the size of building blocks in nanostructured materials as well as the influence of microstructure on physical, chemical and mechanical properties of this material; better understanding of the influence of interfaces on properties of nanostructured material; development of concepts for nanostructured materials and in particular their elaboration, etc.

Nowadays, there is a great deal of interest and activity steered towards extending macroscopic thermodynamics and statistical physics to the nanometer scale consisting of countable particles below the thermodynamic limit due to recent developments in nanoscience and nanotechnology. To generalize thermodynamics on a nanoscale, we should understand well the unique properties of nanosystems. One of the characteristic features of nanosystems is their high surface-to-volume ratio. As the results of surface effects becoming increasingly important with decreasing size, the Gibbs free energy relatively increases for some thermodynamic equilibrium systems. Therefore, the behavior of such nanoscopic clusters differs significantly from the usual thermodynamic limit. It is clearly known that when the system size decreases, one has to consider the fluctuations. Based on the nucleation reactions, the first considerations are given to the temperature fluctuations. The quantitative measurements of temperature fluctuations are realized by superconducting magnetometers. Remarkably that an important role of fluctuations is well underlined in the following statement by the US National Initiative on Nanotechnology: ''There are also many different types of time scales, ranging from 10^{-15} s to several seconds, so consideration must be given to the fact that the particles are actually undergoing fluctuations in time and to the fact that there are uneven size distributions. To provide reliable results, researchers should also consider the relative accuracy appropriate for the space and time scales that are required; however, the cost of accuracy can be high. The temporal scale goes linearly with the number of particles *N*, the spatial scale goes as *O* (*N log N*), yet the accuracy scale can go as high as *N7* to *N!* with a significant prefactor''. Therefore, these valuable hints motivate researchers to pursue the thermodynamic description at the nanometer size for the nucleation of a metastable phase.

For instance, with the help of Raman thermometer the measurements of carbon nanotube temperature within the range 30 ... 250 ºC are made [7]. Those tubes are treated to be standard nanopatterns for testing and calibrating the nanotechnological means. The gained results of experimental research give possibility of realizing the metrologically correct evaluation of temperature measurement results with considering the peculiarities of both measuring instrument and standard pattern. Hereby, to study the action of seven and more possible influence factors (angle of light bunch incidence, distance to a photo-receiver, exposure time, duration of spectrum passing, power and mode of laser functioning, drift characteristics and so on), 28000 gauges have been performed, enabling us to ascertain the following indices of the measurement accuracy. The approach of errors is applied to processing results, consequently of which one of the gained results (with the introduced correction to a systematic error component) looks as T_{real} =287.27 K ± 1.72 K (0.6 %). At the same time, due to the uncertainty approach, the gained result makes T_{real} =287.27 K with the expanded error 0.58 % and combined standard uncertainty 0.3 % at the credence level P = 0.95, the expanded coefficient 1.96 and the efficient value of freedom degrees 130.6 [11].

However, while applying the thermodynamics to the choice of uncorrelated influence factors in the form of thermodynamic coordinates (forces and flows) acting on the thermometric substance of a thermoelectric thermometer, the quantity of necessary measurements is decreased for 1-2 orders, assuring the similar accuracy indices [13]. As a result, the summary influence function in the presence of external thermodynamic fields is determined. Temperature, density, strain and etc. gradients created by the external effect in thermosensitive substance are subordinated to the same statistical regularities as the gradients appearing consequently of a fluctuations effect in the mentioned substance (according to the sense of a fluctuation-dissipation theorem of thermodynamics). At the availability of fluctuations, additional influence functions applying multiplicatively on influence functions related by the fluctuation effect of external environment are formed. The given approach is valuable due to the possibilities of considering the thermodynamic system of thermosensitive substance with regard to external environment, and taking into account the essentiality of fluctuation-dissipation processes that occur in substance itself.

The following nanotechnology development including nanomaterial-lore does not seem to be possible without this in nanometrology [2], whose highly important component is nanothermometry. There are two main issues to consider in nanometrology: precise measurement of sizes within the nanoscope range, and assimilation of existing methods or development of new ones to characterize properties as a function of size and of the certain temperature, pressure and other thermodynamical parameters. Progress in nanothermometry is inseparably related to the nanometrological investigations first of all of nanostructured materials, consequently, to the identification of standard nanopatterns, the performance of their multipurpose and volumetric research and finally to the further development of nanotechnology theoretical principles at electron, quasi-particle, particle and nanothermodynamic decisive levels.

3. Objectives

The study of instrumental facilities of different methods for substance profound structure research on the pattern of thermosensitive thermometer substance in comparison with theoretical developments including the achievements in nanothermodynamics, and so-grounded stipulation of both perspective research methods and theoretical approaches to the further nanothermometry and nanotechnology development (technology+measurement).

4. Theoretical Approaches to Temperature Measurement and Determination in Nanotechnology

Among the considerable number of temperature definitions, one of the fittest for nanothermometry is the following [4]. Temperature is the statistically formed value of quantity, determined by the inner energy of a body of sufficient sizes for the purpose of applying a thermodynamic consideration to this body. So far temperature remains the last and only value among seven main units of International unit system that is still not regulated at the nuclear / molecular and hence much higher level in terms of accuracy. In the chain of leading world centers, the intensive endeavors of elaborating and assuring the unit of temperature scale in the form of a quantum energetic unit at the methodological level of State certifying services are carried out for several years. Applying the direct methods of temperature determination (gas, acoustic, optical, magnetic and noise), and having determined Boltzmann constant with a very small error, we could regulate the unit of a temperature scale due to the energetic units, also endued with a certain determination error. Afterwards, according to the development in nanotechnology, it is supposed to appear the temperature energetic quantum, similar to the quantum of electric resistance, quantum of an electric charge unit, capacitance quantum and so on.

4.1. Principles of Thermodynamic Approach

Dealing with metrology (measurement and result processing), authors have determined the core question - how due to the results of measuring the macro-, micro or just noise-characteristics of the highest level characteristics it is possible to estimate changes at the nanolevel of nanostructured or 1d-, 2d- nanomaterials. To solve it, we should develop the methodology of nanomeasurements as well as principles of interpreting the gained results. Moreover, the approach of I. Prigogine, following which any microscopic process is a result of more or less coherent microscopic processes, should be taken as a basis [15]. Microscopic freedom degrees are revealed as fluctuations describable in terms of the introduced additive members in the equation for microscopic quantities. Exactly in this way we have advanced in the study of drift of a thermoelectric thermometer transformation function. To avoid the correlation effects of different influence factors which are of importance assuring the accuracy in metrology, the metrologically stipulated system of selecting the non-related factors that are thermodynamic quantities is applied [13].

4.2. Nanothermometry, Nanothermodynamics and Temperature of Nanoobjects

Nanothermometry as a branch of nanometrology implies measuring the temperature of nanoobjects with the given accuracy (error, uncertainty), repeatability, discreteness and under the preliminary guaranteed instability of a thermometer transformation function. Moreover, rare works [16] are dedicated to experiencing the term of nanoobject temperature. Almost all known works concern with temperature as one of the main thermodynamic parameters, successfully characterizing substance at the macrolevel. In the light of the stable development of nanotechnology and nanomeasuring, the determination of this task acquires increasing significance, since:

- Single unrepeated gauging is applied whereas there is no place for the classical approach of error theory, evolved in information-measuring systems [17];
- Measuring instrument (sensor) of primary transducer of a measuring means whose interference into energetic exchange disturbs the dimension of the controlled quantity acquires weight;
- Previous selection of certain research tools among the totality of possible instruments capable of retaining the reproducibility of the much needed kind of direct or indirect measurements of a concrete quantity is demanded. Since, when the definition uncertainty is considerable as compared to other uncertainty components, its inclusion into general balance leads to extension of a covering interval [18].

Finally, there appears the task to describe the gained results so that they would satisfy the experiment reproducibility and repeatability as well as the possibility of comparison and verification of the received values of the measured quantity for the purpose of working out its dimension. So in order to determine the temperature of nanoscaled objects, we should:

- Base on the (nano)thermodynamic interpretation of temperature;
- Follow from the existing methods of thermodynamic temperature determination;
- Apply methods connecting the values “temperature” and “controlled parameter”, avoiding the additional coefficients determined with the insufficient accuracy (following other methods), to research the concrete temperature;
- Stipulate the limits and possibilities of applying the notion “thermodynamic temperature” as to the mentioned objects considering their linear sizes;
- Represent the errors of single gauging of “thermodynamic temperature” in hybrid [19] or hybrid-thermodynamic [13] interpretation.

The latter explains the form of systematic component of an instrumental error as an additive totality of multiplicative pairs of influence functions with appropriate coefficients. Forming pairs, where one of the coefficients is determined by fluctuations of thermosensitive substance properties, and another – by fluctuations of parameters of the applied outer fields, corresponds to the sense of fluctuation-dissipation theorem of thermodynamics. Hybrid-thermodynamic approach is used in [18] to evaluate the results of measurement, and its basis concerns with the study of origin sources of errors and particular influence functions. The research of the processes of energy-transmission, based on thermodynamics, enables us to determine a methodical error component as well as cognizable part of systematic component of an instrumental error component, and thus to decrease substantially the guaranteed by the manufacturer of thermometric means a total error of measuring the temperature in exploitation conditions.

4.3. Methodical Errors of Temperature Measurement in Micro- and Nano-world

Avoiding the questions of the gas-medium measurement where owing to the pyrometry means employment there are possibilities of enduing some temperature value through radiation intensity or spectrum to the certain selected atom or molecule, let us consider the methodically more complicated questions of thermometring the solids – micro- and nanoobjects, first of all, their surfaces. The

interactive response of object practically is not figured anywhere consequently of both its own change caused by thermometring and this of thermometer transformation functions during measuring under the effect of energetic-entropy selection. In the conditions of rising significance of energy-transmission processes in the medium "thermometer – controlled object", with decreasing sizes of an object, the intervention of a thermometer into an energetic exchange affects the temperature values, hence causing the occurrence of systematic component of a methodical error component. The more considerable the latter is, the less object we deal with. Therefore, it is proposed for a short term: (a) to create a thermal contact between sensor and object; (b) to conduct the measurements simultaneously; and (c) to determine the temperature and methodical error due to the nature of signal drift.

In the macroworld by default is assumed that linear sensor size does not exceed 0.1-linear size of the controlled object, and the ratio of their volumes – 0.001. This defines a relative methodical error of measurement no higher than 0.1 %. In total, this value loses other components of the measurement error, including the instrumental one. So there are grounds not to consider and not to take into account the methodical error of temperature measurement. For micro-, nano- sized sensors and controlled objects with comparable thermophysical properties, a relative methodical error is $\delta T_{met} = \frac{T_{sen}}{T_x} - 1$. For example, while gauging microobject with T_x = 270 K by means of commensurate-sized sensor of the initial temperature T_{sen} = 300 K, it is received $\delta T_{met} = 11\%$ [5].

4.4 Fluctuation-dissipation Theorem of Thermodynamics and Noise Measurement Methods

Fluctuation-dissipation theorem, relating reversible and irreversible thermodynamics, could be applied both to nanomaterials and sensitive thermometer substance. Nyquist rate, combining the spectral density of energy (SDE) of electric noises within certain frequency band and electric resistance, is treated as a particular case of this theorem. In experimental research, the special attention could be paid exactly to the method of noise investigation both in electric and mechanical treatment. The research in noise- and ultrasonic-thermometry made at the enterprise-designer has assured the high level of metrological reliability of the gained results, and fostered the specification of theoretical states.

The main scientific results of electric noise investigation undertaken on the noise thermometers within the range 4.2 ... 500 K are the following [11]. The known Nyquist formula is equitable for thermal electrical noise at frequencies higher than 1000 Hz and for sensitive elements, made from alloys and composites. Their calibration characteristics are linear, with no recorded deviations. The consideration is important for such an ordinary factor of influence on calibration characteristics as a deformation factor that tends to increase at the combined action of temperature and strains (structural, resilient, ductile or others) in thermosensitive substance during measuring. This factor is present in the local distortions of temperature in the mentioned materials as a result of tensile micro concentrator actions, which are considered as quasi local nanosized defects of thermo-fluctuation origin. The given distortions have lowered substantially the real durability of thermometric substance through electromechanical noises, which at the same time stipulates the deviations of noise thermometer calibration characteristics, worsening its precision. The intensity of electrical noise considerably depends on different transport processes in the thermosensitive substance. For instance, at 77 K, the most significant are the heat and charge transfer processes. At the certain temperatures and deformations, the importance of other processes that may become crucial is growing.

The similar results of characteristics' calibration research in the case of ultrasonic thermometers with tungsten-based half-wave sensitive elements but within the highest temperature range 2000 ... 3000 K are proposed in [10]. Hereby, the transformation function, expressed in a frequency characteristic measured with a minimal error depending on temperature, is caused by direct temperature changes of

Young module. Let us notice that the maximal stability of indices is inherent in sensitive elements made from nanostructured tungsten. The essential role of micro concentrators of mechanical tensions is revealed both in assuring the high-temperature shape-stability with resulting stability of indices, and in appearing *1/f* noise which becomes the pivotal causer of gained bandwidth quality of a thermometer resonator.

Getting into a tungsten matrix, admixture and addition atoms reveal a deformation influence depending on the ratio of their atom volumes and volume of a main component. For instance, the ratio of atom volumes of potassium – main admixture of silicon-lixivium addition – and tungsten makes near 5 : 1. Consequently, mechanical tension fields, made by addition atoms, influence upon the state of substances and finally affect an USTh transformation function. Admixtures of potassium, indissoluble at the room temperatures in a tungsten matrix, are concentrated in the form of liquid phase bubbles. With rising the temperature till 1750-1850 K, they evaporate forming micropores inside of which the pressure reaches 30-40 MPa. As a result, the field of mechanical tensile nanoconcentrators is being formed in a matrix, stabilizing a structure. Their influence could be taken into consideration in terms of consolidation theory of porous metal ceramic materials. Hereby, mechanical tensions are applied exactly to pores without affecting monolith. Therefore the changes of an USTh transformation function could be related through Young module to the alteration in porosity, and correspondently through the prefactor ρ/ρ_0 – to that in substance density from ρ_0 to ρ: $\frac{\Delta f(T)}{f} = \frac{1}{2}\left(\frac{\rho}{\rho_0}\frac{\Delta E_U(T)}{E_U(T)}\right)$.

5. Following Development of Theoretical Approaches and Experimental Methods

Up to date, there are two kinds of fundamental approaches to open out in thermodynamics on nanoscale, based on the microscopic and macroscopic viewpoints, respectively. One would go back to the fundamental theorem of macroscopic thermodynamics and establish the new formalism of nanothermodynamics by introducing the new function(s) presenting the fluctuations or surface effects of nanosystems. Another one could directly modify the equations of the macroscopic thermodynamics and establish the new model of thermodynamics on nanoscale by incorporating the Laplace–Young or Gibbs–Thomson relations, representing the density fluctuation of nanosystems in the corresponding thermodynamic expressions [3, p.89]. Let us apply both of mentioned approaches to the description of thermometer behavior and metrological characteristics incl. sensitive elements made from nanoscaled and nanostructured materials.

5.1. Thermodynamic Quantities and New Formalism of Nanothermodynamics while Describing the Peculiarities of Thermometer Exploitation

5.1.1. Nanothermometers with Solid- and Liquid-phase Sensitive Elements

Thermometers with liquid- and solid-phase sensitive elements are already well-known in thermometry [9]. Their construction implies the availability of a narrow tube with movable thermosensitive substance inside. The consideration given to a liquid thermometer in macroworld under the condition of neglecting superficial tension, when the diameter of a thermometric tube is quite large and thermosensitive substance is hardly contractile, could bring in the analysis of the equation describing the interrelation of the volume of the mentioned substance and the changes of column sizes Δh of a thermometer with its temperature: $\Delta h = c\, d \Delta T$, mm **(***d* - the inner diameter of a thermometric tube; *c* – constant**).** Hereby, the sensitivity of a thermometer is reducing with the decrease in the diameter of a thermometric tube. Here the effect of superficial tension forces is revealed only through the distortion of meniscus causing a readout error.

The similar equation is deduced also for micro- and nanothermometers filled with solid-phase thermosensitive substance. These sensitive elements that function due to the phenomenon of thermo expanding are principally similar to the sensitive elements of liquid thermometers of macrosizes. The size of the employed micro- and nanotube is being reduced while bringing thermometer dimensions onto micro- and afterwards nanoscale. Modern investigations cover the application of single-wall carbon nanotubes with the diameter approximately 10 ... 40 nm.

In the case of a nanothermometer with a liquid-phase sensitive element**,** the processes of transposition related to the superficial freedom degree prevail. The behavior of liquid in a capillary tube depends on the phenomenon of moistening. The temperature-dependence of superficial tension could be depicted for all liquids so that the data are placed along one common curve: $\sigma V^{2/3} = k\left(T_c - 6 - T\right)$. Here, $k = 2.1\times10^{-7}$ Joule/K mole$^{-2/3}$ – Eötvös constant that is equal for all liquids. The equation of calibration characteristic of a nanothermometer with a liquid-phase sensitive element could be found as $\Delta h = b\left(T_c - 6 - T\right)/d$ (b – constant).

Evidently, the constant of calibration characteristic of a nanothermometer with a liquid-phase sensitive element as well as the constant of this of a nanothermometer with a solid-phase sensitive element comprises the size of a determinant constructive element, namely the inner diameter of the thermometric tube d. However, the main difference between them is that the sensitivity of a nanothermometer with a liquid thermosensitive element rises relatively to the decrease in a tube diameter, whereas the sensitivity of a nanothermometer with a solid-phase sensitive element falls [9]. Therefore, in the latter case, we should orient not to readings through solid-phase sensitive substance, but due to the dimension of an air column above.

5.1.2. Resistance Thermometers with Nanostructured Thermosensitive Substance

Metal glasses (MG) with an amorphous structure undergo our investigations as the new-gained materials with the high specific resistance ρ at the small value of its temperature coefficient α. 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 evolving the special electrotechnical materials, spintronics [20] and etc. The study of nanostructured materials confirms the existence of fields of considerable mechanical microtensions whose influence is equal to the doping with a number of admixtures. In this case, we have applied nanothermodynamics [3] in order to explain MG electrokinetics 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. Considering the two-phase MG model, i.e., assuming that there are precipitations of other phases in a matrix, e.g. pseudo-phases which could be represented by the microvolumes with different densities, to obtain experimental results, we evolve the equation of the third order for the specific electrical resistance of two-phase material: $\rho_a\left(\Delta\rho\right)^2 = \left(1-q\right)\rho_1\left(\Delta\rho\right)^2 + \left(\Delta\rho\right)^3 - q\rho_1^2$. Here $q = \Delta S/S$ - an efficient intersection area of precipitations, $\rho_1 = a + bT$ - specific electrical resistance of a matrix, $\Delta\rho$ – changes of the specific electrical resistance due to precipitations. Explanation roots in the dependence of the specific volume 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 [12].

5.1.3. Thermoelectric Thermometers, Thermometric Substance and its Structure

Phenomenology [9] of thermo-e.m.f. stability of mechanically strained (σ - tension) thermometric material has ascertained the fact of Gibbs energy σ^2/E_U amplification and enabled us to stipulate the introduction of additional thermodynamic power $\overline{X}$, caused by the drop in this energy: $\overline{X} = \nabla\left(\sigma^2/2E_U\right) = \sigma/E_U \nabla\sigma$. For better understanding of processes that happen in materials, thermometer drift is related to the temperature change in small substance volumes (with or without deformation). In thermosensitive substance, the notified below hierarchy of structural levels with regard to the deformation/distraction processes is observed.

The structural levels, utmost by their scales, are treated to be stiffly given. The first of them is a macroscopic level, describing the chain of defects with the help of a distribution function, smoothly changing due to volume. In thermometry it is realized by introduction of dependable on temperature, distributed physical properties of thermometric substance, or parameters of classical thermodynamics which helps to reach the change in (thermo)electric properties. In [21] (Fig. 1), the temperature changes of solid bodies, depending on the applied efforts (an adiabatic case) in a tense-deformation state, have been thoroughly studied. The temperature fall is fixed in the resilient deformation area. With crossing into the area of ductile deformation, temperature changes in local volumes of the deformed substance get the tendency to change a sign.

Intermediate area of a mezoscopic level (10^{-8} – 10^{-5} m) is placed between macroscopic and nanolevel which concerns with the certain lattice defects (main physical processes – kinematics of dot defects, diffusion ductility and correspondent changes of thermoelectric properties). In this area, the processes of precritical evolution of the deformed material take place. Hereby, the considerable tension gradients are being formed. In the opinion of [22] under the conditions of relative pattern prolongation 10^{-4} as an issue of residual viscous deformation, a deformation potential makes 0,5 mV which affects thermoelectric properties, particularly, thermoelectric heterogeneity. When the sizes of certain crystals become commensurable with the characteristic size (wire diameter) of recrystallized material, the level of mechanical tensions on the border of non-recrystallized and recrystallized areas reaches 10^3 MPa, which leads to linear changes in thermo-e.m.f.

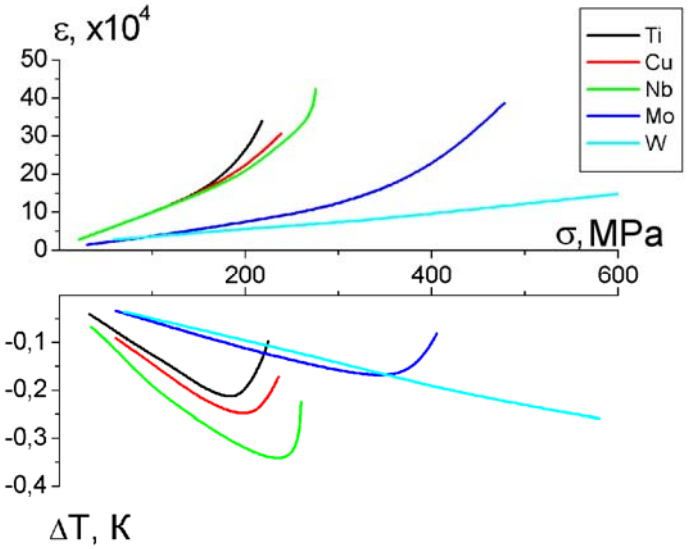


Fig. 1. Correlation of mechanical tensions and deformations in resilient area with pattern temperature change, fixed by means of a thermoelectric thermometer [21].

The second determinable level is a nanoscopic level. The decrease in the real limit of material strength concerning the theoretical values is explained by the presence of tensile defects [23], [24]. The latter are considered as micro- or nano- concentrators of mechanical tensions, according to the results of analyzing the spectrum of combinational light in the transparent polyethylene terephthalate. The availability of low- and high-frequency satellites with reference to the main maximum 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 inter-atomic bonds of tensile defects for the value $\varepsilon = -\frac{1}{G}\frac{\Delta\nu}{\nu}$. Here G - the mode parameter of Gruneisen, $\Delta\nu$ - the alteration of light frequency, due to multi-phonon dispersion.

While capturing the phonons by a tensile defect, its local temperature is rising. Considerable thermo-extension takes place and hence at some moment could cause the rapture of inter-atomic bonds and thus microcrack appearance. Using the intensity of fluctuation bands in Raman spectrum of combinational dispersion (Stokes and Anti-Stokes bands), the phonon value or the capture by tensile defects was ascertained. Local temperature of the above mentioned defects (with linear size till 10 atoms) is determined as $T_d = \frac{h\nu}{k\left[\ln(n+1) - \ln n\right]}$. It is obvious how essential the dot temperature increasing is on these defects, being especially important for the thermosensitive substance.

Microconcentrators serve as a trigger that launches the transformation of the energy applied to the substance 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 defects as physically elementary nanosized structure subsystems enables us to motivate the introduction of thermodynamic values i.e. the application of nanothermodynamics of irreversible processes to the researched substance.

Since metal glasses of *Fe-(Ni)-B* system, whose structure resembles unordered liquid, and a deformation nature is viscous, are referred to nanostructured materials, we have studied temperature-mechanical factors' effect on the changes in calibration characteristics of thermoelectric thermometers. The high reproducibility of characteristics and their negligible drift (till 11 nV) at high-temperature maturing under the substantial extension efforts (Fig. 2) is noted. It proves [12] the determinable role in the drift appearance of both a mechanical tension gradient and concentrators of mechanical tensions.

5.2. New Model of Thermodynamics on Nanoscale with Incorporating the Real Defect Structure

In the concrete case of thermometric substance, thermodynamic methods do not allow ascertaining the form of an equation of a thermodynamic system state, required to endow the equation of thermodynamics with necessary physical sense. On contrary, this determination takes place owing to statistic physics as an indisputable thermodynamic component predicting the necessity of fluctuation presence [25]. Therefore considering the problems of noise inherent in a solid state and originated by thermo- or $1/f$ low-frequency, let us plunge into the detailed peculiarities of electron-phonon interaction, applying different approximations on the subject of possible ways of their adequate description by statistic physics. For a solid body in the case of possible Hooke's law applying, the normal lattice fluctuations are treated to be independent. The energy of these fluctuations is determined by their frequency ω, and depends on the quantum number n of phonon states. In thermo-equilibrium, the mean value of the quantum number n is found by Planck law, representing the function of phonon distribution through the frequencies [26]. Energy (of acoustic or optical modes) of phonons could be approximated by the Debye model based on Planck's law at $\hbar\omega << kT$.

The Einstein model applies the same formula while assuming that $\hbar\omega >> kT$. However, no model describes intermediate, in our opinion, the real state of a solid body when phonon energy becomes commensurable with *kT*, and in the body structure, heterogeneities of nano-scaled tensile quasi-defects capable of modifying the very process of electron-phonon interaction appear. The presence of the latter [23] implies, at first, that the consideration is not made at the level of linear oscillators but physically elementary substance volumes in whose case thermodynamic consideration already has a sense; at second, that there exists ambiguity in a substance structuring nature at the atom-electron level as well as the appropriate ways of its study during electron-phonon interaction (through SDE while studying electric noise, or the spectral distribution of an inner friction parameter while studying the latter).

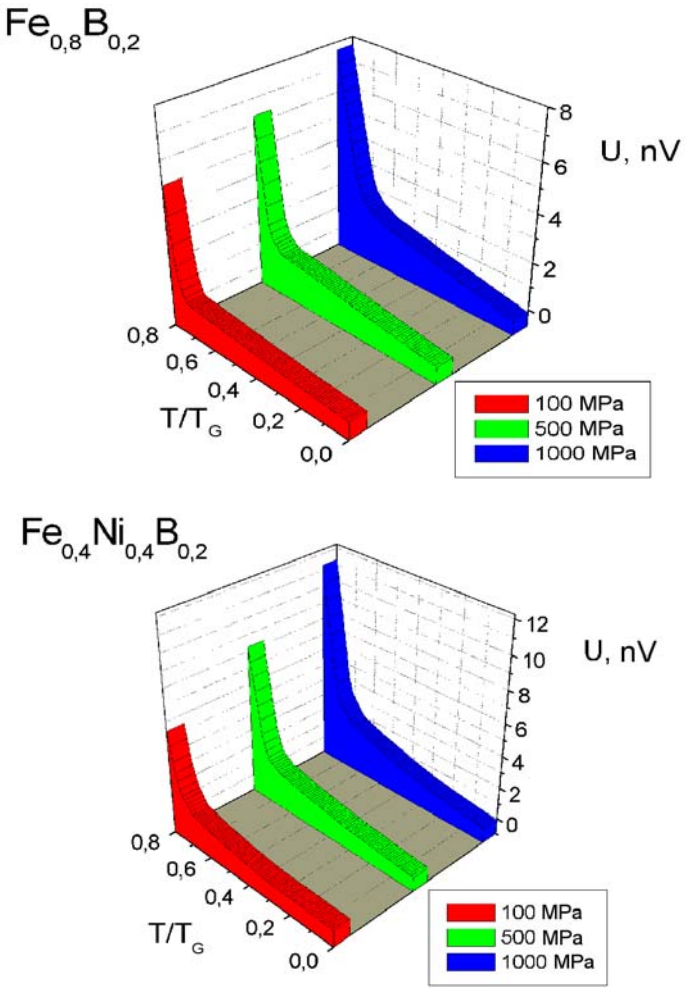


Fig. 2. Temperature dependence of changes in integral thermo-e.m.f. of metal glasses at the different extension efforts.

Let us accept that phonon energy is insignificant: $\hbar\omega < kT$. It means that the situation could be liable to the description through the Debye model with a distribution function that is reduced to $\langle n\rangle = \frac{kT}{\hbar\omega}$ (the symbol $\langle\rangle$ means a mean value in thermo-equilibrium). To wit, at the attenuated process energetics when the speed of energy incoming cedes the speed of its removal at any current moment of time, the given defect acts as a virtual trap of phonons. Having absorbed a phonon and received an energy quantum, quasi-effect immediately gets rid of the latter. However, the similar methods are not capable of describing the system behavior revealed in a certain set of researched physical properties. Thus, at n' synchronously acquired phonons on one tensile defect (multi-phonon dispersion, fixed e.g.

by Raman method [27]), Planck law describes no longer the state of thermo-equilibrium in the whole volume of substance, but that of physically elementary part of the mentioned volume relevant to the concrete defect:

$$\langle n_d \rangle = \frac{1}{e^{\frac{n'\hbar\omega}{kT}} - 1} \tag{1}$$

Defects with temperatures over a thousand degrees are fixed in [24] at spectroscopic study of transparent polymers during deformation at room temperature. Then at $n'\hbar\omega > kT$, i. e. at multi-phonon dispersion, the total energy of phonons essentially exceeds *kT*, causing the passing to the Einstein model with the distribution:

$$\langle n_d \rangle = \exp\left(-\frac{n'\hbar\omega}{kT}\right) \tag{2}$$

It does not affect the previously determined limits of the mentioned models. As we discuss not so the ordinary oscillators but rather the fluctuation oscillators of a nanolevel, whose usage helps to explain the lowering of a material robustness limit in comparison with theoretical values. At rising the total energy of phonons, accumulated on a defect, the latter finishes its role of an energetic concentrator with its following conversion into a microcrack. It happens when the speed of energy incoming exceeds that of its removal [28].

On the other hand, in the thermometric substance of an ultrasonic thermometer, the possibilities of such physically elementary volumes to absorb the energy of an elastic-plastic wave, are characterized by e. g. the absorption factor $\Pi = \frac{\Delta W}{2\pi W}$, here ΔW is a part of the common wave energy *W* absorbed by the body. In the case of the Debye model, the absorption factor is expressed through the frequency: $\Pi = \frac{\Delta\omega}{2\pi\omega} = \frac{\Delta f}{2\pi f}$. If it is reduced to the single frequency range Δf, then it corresponds to the spectral absorption factor:

$$\Pi_\omega(f) = \frac{\Pi}{\Delta f} = \frac{1}{2\pi f} \tag{3}$$

In the case of the signals of smaller energy (lower frequency), we could treat the medium response to be linear. Then fluctuation SDE is proportional to the spectral absorption factor:

$$S_\omega(f) = k_p \Pi_\omega(f) = \frac{k_p}{2\pi f}, \tag{4}$$

here k_p - the coefficient of power transmission of a measuring system. To wit, consequently of the justified Debye model application, the frequency SDE dependence inherent in *1/f* noise is gained.

In the case of the Einstein model usage, at the concentrating of energy phonons on the physically elementary volumes, tensile quasi-defects, we come to the determination of an absorption factor as:

$$\Pi = \frac{n'\hbar\Delta\omega}{2\pi kT} = \frac{n'\hbar\Delta f}{kT} \tag{5}$$

and a spectral absorption factor being proportional to $S_\omega(f)$:

$$\Pi_\omega(f) = \frac{\Pi}{\Delta f} = \frac{n'\hbar}{kT} \tag{6}$$

It is frequency independent, and fitted for the case of thermal noises. The experimentally fixed quadratic SDE nature of *1/f* noise could be stipulated by a methodical measurement error, and a higher order of degree dependence, till cubic, obviously, is related to the restriction of frequency-time analysis range and to the integration of the gained signal whilst gauging the substance remains in non-stationary non-equilibrium thermodynamic state [29].

6. Conclusions

Development of research methodology with advancing in the theory and practice of both measurement error and uncertainty approaches enables us, at first, to describe micro- and nanoobjects characteristics in a more precise and reproductive manner and, at second, to deepen the microworld insight, assuring the development of nanotechnology. Nanometrologically and nanothermometrically provided reproducibility of loop turns to be of exceptional significance in nanopattern production. The combination of metrological and especially thermometric methodologies with theoretical research while using and further unifying the gained data requires the proper understanding of nanostructures in order to reinforce our conviction that the revealed "artifacts" are not created in the very information-measurement system. Hereby, the optimization of measurement quantity, providing the proper metrological characteristics, should comprise the elimination of correlation effects. This is carried out by the qualified selection and estimation of influence factors whose independence is conditioned by means of nanothermodynamics.

Since the sizes play a crucial role in ascertaining the properties of nanomaterials, and new phenomena [2] are observed in nanopatterns, some of them being important for nanothermometry are considered in detail:

- The intensive atom diffusion across the division surfaces of thermosensitive substance, involved in the Laplace–Young or Gibbs–Thomson relations;
- The decrease in a temperature coefficient of electric resistance in metal glasses of resistance thermometers till zero, caused by the precipitations of the second phase;
- Immediate action of superficial tension forces (the main component of the state equation of nanothermodynamics), regarded in forming the calibration characteristics of capillary type thermometers at decreasing their sizes into a nanoarea and etc.

Hereby, the issues of influence of the change in a phonon spectrum, so-called effect of phonon confinement, are of primary significance in forming the calibration characteristics of Raman, ultrasonic and noise thermotransducers. Particularly, this effect explains the appearance of *1/f* noise and its transformation into the thermal noise that has a special sense in nanothermometry and eventually in nanometrology.

Thermodynamically stipulated phase equilibrium is replaced due to the contribution of division surfaces or superficially predetermined mechanical tensions to free energy of thermosensitive substance system which enables us to produce new quasi-nonequilibrium materials with a high stability of calibrating characteristics for thermoelectric thermotransducers, and also to create functionally gradient thermocouples that are a bright example of structures, quasi-distributed in space [14].

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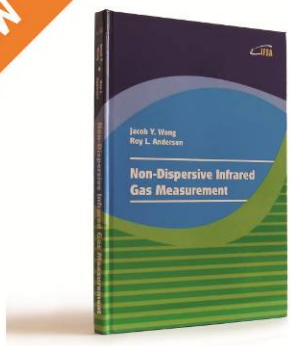
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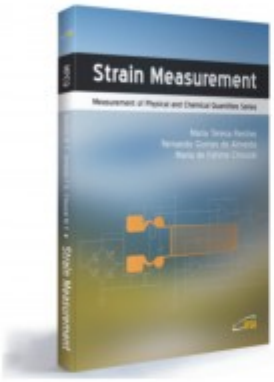
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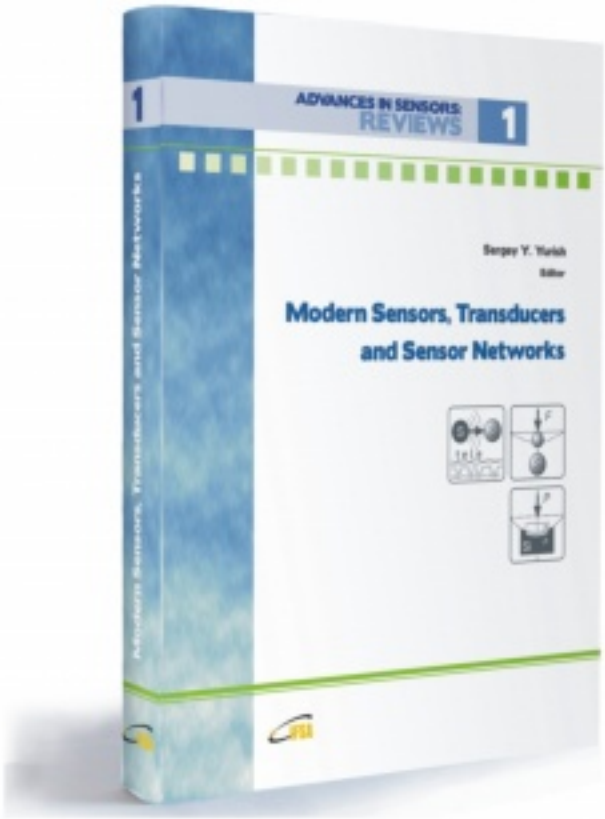
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