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METHODOLOGICAL APPROACH TO THE DIMENSION ESTIMATION OF MODIFYING PARTICLES FOR NANOCOMPOSITES

Abstract: The theoretical basis of the methodological approach to the analytical form for evaluation of the dimension boundary L_0 between nano- and macro-states of condensed matter is considered. The feasibility for using the Debye characteristic temperature, pulse energy, frequency and wavelength to determine the nanoscale dimension of particles with different composition is shown. It is shown that the proposed formula $L_0 = C \theta_D^{-1/2}$ reflects the effect of particle size on the parameters of the characteristics of their physical properties and can be used for nanocomposites modifiers.

Key words: nanostate, nanoparticle, size boundary, Debye temperature, dynamic models.

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Introduction

Nanocomposite materials based on polymer, metal, ceramic, and other matrices are increasingly used in various branches of industrial production. Moreover, the formation of the structure of such composites is determined mainly by the parameters of nanoscale components, which are introduced into the

matrix using various technological methods [1–3]. According to established practice, a nanocomposite is defined as “a multi-component solid material in which one of the components in one, two or three dimensions has sizes not exceeding 100 nm” (Nano-composite Science and Technology, Wiley-VCH, 2003). At the same time, studies of the nanostate phenomenon

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indicate the ambiguity of the prevailing approaches to determining the dimensional parameters of nanoparticles used in various technical applications.

In previously published works [4–11, 24], a formula was proposed that establishes the relationship between the size boundary and the nano- and macro states (L_0) with the Debye temperature (θ_D). This temperature is the boundary between the applicability of the corresponding physical theories to description the properties of substances at various temperatures (T). At $T \gg \theta_D$, the phonon spectrum contains both low- and high-frequency vibrational modes. At $T \ll \theta_D$, there are no high-frequency modes. These two areas are usually called classical and quantum. In classical – the law of Dulong and Petit is applicable, in quantum – the heat capacity at constant pressure monotonously decreases with decreasing temperature. The classical and quantum approaches are the more reliable, the stronger the inequalities $T > \theta_D$ or $T < \theta_D$ are. But a certain effect on the spectral composition of lattice vibrations can also be exerted by the size of the sample. Therefore, when considering phonon parameters, a number of studies indicate that the analysis of the frequency distribution refers to the samples under study with large volumes, i.e., the influence of dimensional factors on the parameters of the characteristics of the sample is excluded.

At the same time, numerous literature data (for example, [12–17]) indicate that, in the nanoscale range (in the nanophase), the parameters of the characteristics of a substance particle significantly differ from similar parameters for objects with significantly larger, compared atomic size (macrophase).

The aim of this work is to consider methods for estimating the particle size parameters characterizing a nanostate using the modern concepts of condensed matter physics, quantum physics, and nanomaterial science.

Results and discussion

According to the Einstein and Debye dynamic models of crystals, atoms in condensed matter oscillate near their ideal, that is, equilibrium, positions. Therefore, a crystal can be represented as a system of oscillators interacting with each other. Obviously, all atoms in such a single-element structure are in the same crystal-physical positions and, therefore, having studied the state of any of them, we can characterize the state of the whole system. In this model, the atoms are as if connected to each other by quasi-elastic force, and the displacements from the equilibrium position are not very large, that is, we can assume that all atoms oscillate with the same frequency ω . It is precisely on this assumption that Einstein's theory is based, which made it possible to explain the deviations of the specific heat parameter of substances at low temperatures from the Dulong and Petit law [18–20].

Einstein believed that each of the N atoms has 3 degrees of freedom, i.e., there are $3N$ independent harmonic oscillators in the crystal, oscillating with the same frequency ω and having energy $E = \hbar\omega$ (\hbar is the Planck constant). The principles of quantization allow the presence of states with energy

$$E = (n + 0,5) \hbar\omega, \quad (1)$$

where n is an integer; zero-point vibrations are taken into account by the term $0.5 \hbar\omega$.

Oscillations of atoms in crystals, as in any condensed medium, lead to the fact that they generate waves of mechanical excitation, which, as follows from equation (1), are described by quanta of mechanical excitation, called phonons.

At low temperatures, the natural phonon frequency can be such that the inequality $\hbar\omega_j \gg kT$ holds. In this case, $\exp(\hbar\omega / kT) \gg 1$.

At the same time, the contribution of these components to the total energy of the system will be insignificant, since their number in the frequency spectrum is small [18]. Therefore, we can restrict ourselves to the acoustic branches of phonons. The average energy in this case is

$$\bar{E} = \hbar\omega \exp[-\hbar\omega / kT], \quad (2)$$

that is, the heat capacity at low temperatures ($T \rightarrow 0$) is determined by the condition:

$$C_v = Nk \left(\frac{\hbar\omega}{kT} \right)^2 \exp[-\hbar\omega / kT]. \quad (3)$$

It follows that $C_v \rightarrow 0$ as $T \rightarrow 0$ according to the law $\exp(-\hbar\omega/kT)$.

Since all oscillators in the Einstein model have the same frequency (ω), i.e., the same energy, the temperature factor θ_E of the form introduced:

$$\hbar\omega = k \cdot \theta_E, \quad (4)$$

where θ_E is the Einstein temperature.

In this case, the heat capacity (3), taking into account (4), at $T \rightarrow 0$ takes the form:

$$C_v = Nk \left(\frac{\theta_E}{T} \right)^2 \cdot e^{-\theta_E/T}. \quad (5)$$

For a more rigorous assessment of the dependence of C_v on T at low temperatures, Debye abandoned the model of identical atomic oscillators, and suggested that atoms in a solid oscillate with different frequencies.

The frequency distribution of the oscillators is described by the dependence $P(\omega)$, which is determined by the conditions [18–20]:

$$P(\omega) d\omega = \frac{3\omega^2}{2\pi^2 v^3} d\omega = C\omega^2 d\omega, \quad (6)$$

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where v is phonon propagation velocity.

In accordance with Debye assumption, the frequency ω can reach a certain maximum value $\omega_m = \omega_D$, which is called the Debye frequency.

With the frequency distribution function (6), the internal energy density is:

$$(U - U_0) = \frac{3\hbar}{2\pi^2 v^3} \int \frac{\omega^3}{e^{kT} - 1} d\omega = \frac{3k^4 T^4}{2\pi^2 \hbar^3} \int_0^{x_m} \frac{x^3}{e^x - 1} dx, \quad (7)$$

where U_0 is the energy of the equilibrium state of the lattice.

The integration variable x in (7) varies in the range from 0 to x_m , which is equal to

$$x_m = \frac{\hbar\omega_m}{kT} = \frac{\hbar q_m c}{kT} = \frac{\hbar v (6\pi^2 n)^{1/3}}{k} \cdot \frac{1}{T} = \frac{\theta_D}{T}, \quad (8)$$

where n is the number of atoms per unit volume (atomic density), q_m is the wave number, c is the speed of sound [18-20]. The quantity θ_D is related to the Debye frequency by the equation

$$\hbar\omega_D = k\theta_D. \quad (9)$$

If the dependence of C_v on T in the Einstein approximation is exponential, then in the Debye approximation $C \sim T^3$, which is more consistent with the experiment. Therefore, Debye's idea can be considered reasonable that when the values of the phonon frequency modes go to the region $\omega > \omega_D$ under the influence of certain factors, the mechanism of the physical processes will change compared to the case when there are only frequencies $\omega \leq \omega_D$ [18].

The Debye model, like the Einstein model, explains the experimentally discovered fact of a decrease in heat capacity at a constant volume (C_v) at temperatures below the characteristic temperature.

The principle of frequency distribution is applicable to describe samples with sufficiently large volumes of substances. In this case, the sample size does not affect the dependence $P(\omega)$. It follows from the experimental data that there are two regions: the low-frequency branch $\omega < \omega_D$ and the high-frequency branch $\omega > \omega_D$ in the frequency spectrum of harmonic oscillators of a substance particle.

If the size of the crystal (particle) is sufficiently small, then the formation of phonons with long wavelengths is impossible, that is, the size of the crystal "cuts off" the acoustic, low-frequency branch in the spectrum. Provided that the minimum phonon frequency exceeds ω_D , the processes occurring in the sample differ from similar processes in samples with large geometric sizes. Consequently, if the sample size L is such that low-frequency modes with a wavelength longer than L cannot appear in it, then physical processes in this sample will proceed differently than in a large sample of the same substance at the same temperature. Since in this case L_0 corresponds to the condition $\lambda_D = v/\omega_D$ (v is the phonon propagation velocity), we can draw the following conclusion: for

each substance, there is a boundary dimension L_0 related to the Debye temperature θ_D . If the sample size is $r > L_0$, then this sample has the properties of large samples, if $r < L_0$, then the size factors influence the properties of the sample, i.e., with a further decrease in the particle size, excitations with a Debye frequency are impossible in them, since the Debye wavelength does not "fit" between the surfaces of the sample. The value of the parameter L_0 is different for substances of different composition and structure. For example, the parameter L_0 is 8 nm for diamond, and 30 nm – for rubidium. As can be seen, for the studied substances, the parameter L_0 does not correspond to the value of 100 nm [21, 22].

The influence of the size factor on the nature of the physical processes in different substances was considered in [1, 2, 23]. It was found that the parameters of optical, magnetic, and other characteristics of particles substantially depend on size.

Let us consider various approaches to the determination of the L_0 parameter.

I. It is known from the classical concepts of quantum physics, that mechanical excitations in the crystal lattice are carried out by electronic processes, that is, the interaction between atoms is due to the interaction of their electrons [18 – 20, 22].

The electron wave function is an eigenfunction of the momentum operator:

$$\hat{P}\psi = P\psi, \quad (10)$$

moreover, the momentum operator has the form:

$$\hat{P} = -i\hbar \sum_{i=1}^3 (x_0)_j \frac{\partial}{\partial x_j}, \quad (11)$$

where $(x_0)_j$ are the unit vectors of the coordinate axes $x_1 = x, x_2 = y, x_3 = z$.

Let us consider the case when $P_x = P_y = P_z$, that is, an isotropic substance is analyzed. Then equation (10) takes the form:

$$\frac{\partial \psi}{\partial x} + \frac{i}{\hbar} \cdot P_x \psi = 0. \quad (12)$$

The solution to equation (12) is as follows:

$$\psi = \psi_0 \exp\left(-\frac{i}{\hbar} P_x \cdot x\right), \quad (13)$$

i.e., the maxima of the real part of the wave function ψ correspond to the condition

$$P_x \cdot x = 2\pi\hbar \cdot n = hn, \quad (14)$$

where n is an integer.

Let us consider the smallest pulse value when $n = 1$ in equation (14). The lattice energy corresponding to the smallest pulse value is taken equal to $E = E_D$. Since the Debye energy is equal to

$$E_D = \hbar\omega_D = k\theta_D = \frac{3(P_x^2)_D}{2m_e}, \quad (15)$$

then taking into account (14) and (15), we'll get

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$$P_x \cdot x = \left(\frac{2m_e \cdot k\theta_D}{3} \right)^{1/2} \cdot x = h. \quad (16)$$

Since we consider the momentum corresponding to the Debye temperature, the size x will be equal to L_0 , where L_0 is the dimensional boundary between the macrostate and nanostate. In view of the above, it follows from equations (13 – 16):

$$L_0 = \frac{\sqrt{1,5} h}{\sqrt{m_e \cdot k}} (\theta_D)^{-1/2} = C (\theta_D)^{-1/2}. \quad (17)$$

The physical constants in (17) have values: $h = 6,63 \cdot 10^{-34}$ J·s; $k = 1,38 \cdot 10^{-23}$ J·K; $m = 9,11 \cdot 10^{-21}$ kg, that is, the value of the constant C in formula (17) is $230 \text{ nm} \cdot \text{K}^{-1/2}$. Hence

$$L_0 = 230 \cdot (\theta_D)^{-1/2} [HM]. \quad (18)$$

II. Consider the possibility of deriving formula (17) based on other approaches [22].

Debye temperature (θ_D) determines both Debye energy (E_D) and Debye momentum (P_D):

$$E_D = k\theta_D \quad (19)$$

$$P_D = \left[\sum_i P_i^2 \right]^{1/2} = (2E_D \cdot m)^{1/2}, \quad (20)$$

where i is the index of the coordinate axis (x, y, z).

The Debye momentum is a boundary value for using the classical and quantum approximations. When passing to small-sized crystals (particles), the nature of phonons distribution will differ from the process in the macroparticles. Obviously, phonons with a wavelength of $\lambda > 2l$ (l is the particle size) cannot exist. Applying the uncertainty relation taking into account the fact that $P = P_D$, we obtain the particle size $l = L_0$ at which the change in the properties of the substance occurs, that is, the particle acquires the characteristics parameters due to dimensional factors. Hence:

$$P_D \cdot L_0 = h. \quad (21)$$

Substituting in the equation (21) the value of the Debye momentum (20) along one of the coordinate axes, we obtain:

$$P_i \left(\frac{2k\theta_D m}{3} \right)^{1/2} \cdot L_0 = h. \quad (22)$$

From here we get:

$$L_0 = \frac{h\sqrt{1,5}}{\sqrt{km}} (\theta_D)^{-1/2} = C (\theta_D)^{-1/2}. \quad (23)$$

The analytical expression for estimating the parameter L_0 of nanoscale particles of material substances include the Debye temperature (θ_D). Debye temperature is a relatively conditional parameter characterizing a substance, since its determination involves the use of a number of approximations. This parameter is tabulated and widely used in the physics of condensed matter. Debye temperature (θ_D) is determined from the condition:

$$k\theta_D = h\omega_D = h\bar{v} (6\pi^2 n)^{1/3}, \quad (24)$$

where n is the average density of atoms (the number of atoms per unit volume); \bar{v} is the average speed of sound in a substance; ω_D is the limiting frequency of elastic vibrations.

The parameter L_0 is an anisotropic quantity, i.e., the limiting size characterizing the nanostate can differ for different directions.

An experimental assessment of the adequacy of the obtained expression was carried out in the analysis of particle size parameters of various compositions and structures. The calculated values of the maximum sizes of nanoparticles are given in Tables 1 and 2.

Table 1 shows the values of Debye temperature (θ_D) and the values of the dimensional boundaries between nano- and macrophases calculated using formula (18). The indicated parameters are located as the Debye temperature increases. The values of the parameter θ_D are taken from various literary sources. If the values of θ_D are the same or are found only in one of the sources, then a single value is given, if different – then the maximum and minimum values are indicated. In this case, the experimentally established fact of the stability of the θ_D parameter in a wide temperature range was taken into account [25, 26]. The limiting values of the sizes of nanocrystals were similarly determined.

Table 1 – Characteristic temperatures (θ_D , K) and calculated maximum nanocrystal sizes (L_0 , nm) of some substances

№	1	2	3	4	5	6	7	8
Substance	Ne	Pr	Ar	Tl	Pb	Hg	K	In
θ_D	63	74	85	89-96	88-94,5	60-100	100	109-129
L_0	29,0	26,7	25,0	24,0-23,5	24,5-23,6	29,7-23,0	23,0	22,0-20,3

9	10	11	12	13	14	15	16	17
Bi	La	Gd	Na	Au	Sn (white)	Sb	Sn (grey)	Ag
117-120	132	152	150-165	168	170-189	200	200-212	215-225
21,2	20,0	18,7	18,8-18,0	17,7	17,7-16,7	16,3	16,3-15,8	15,7-15,3

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18	19	20	21	22	23	24	25	26	27	28
Ca	Pt	Ta	Zn	Ga	Nb	Zr	V	Pd	Ti	As
219-230	229	231	234-308	240	252	270	273	275	278	285
15,5-15,2	15,2	15,1	15,0-13,1	14,8	14,5	14,0	13,9	13,8	13,8	13,6

29	30	31	32	33	34	35	36	37
Cd	W	Ge	Mg	Cu	Ni	Co	Mn	Li
220-300	270-379	366	318-406	315-445	375-456	385	400	400
15,6-13,3	14,0-11,8	12,0	12,9-11,4	12,9-10,9	11,9-10,8	11,7	11,7	11,5

38	39	40	41	42	43	44	45
Mo	Al	Cr	Fe	Si	Be	B	C(алмаз)
380-425	294-418	402-460	420-467	625-658	1000-1160	1250	1850
11,8-11,1	11,6-11,2	11,5-10,7	11,2-10,6	9,2-9,0	7,3-6,8	6,5	5,3

Table 2 shows the characteristic temperatures and determined by formula (18) limiting sizes of nanocrystals of some halides.

Table 2 – Characteristic temperatures (θ_D , K) and maximum sizes of nanocrystals (nanoparticles) (L_0 , nm) of some halides

№	1	2	3	4	5	6	7
Substance	RbI	KI	RbBr	AgBr	NaI	RbCl	KBr
θ_D	103	131	131	150	154	165	173
L_0	22,7	20,1	20,1	18,8	18,5	18,0	17,5

8	9	10	11	12	13	14
NaBr	KCl	NaCl	KF	LiCl	NaF	LiF
224	231	320	336	422	492	730
15,4	15,1	12,8	12,5	11,2	10,4	8,5

One of the objections of the applicability the Debye temperature parameter for calculating the size boundary L_0 between the nano- and bulk states is that for nanoparticles it can differ significantly from the similar parameter characteristic of the bulk phase. An analysis of the literature data presented in [22] shows that the value of the parameter θ_D does indeed change but increases slightly compared with the values characteristic of the bulk phase. Therefore, its use for calculating the parameter L_0 can be considered reasonable.

Thus, the use of the obtained analytical expression for determining the limiting size of a nanoparticle selected for modifying the polymer matrix is correct, and the calculated values of the parameter L_0 can be used to select the components of nanocomposite materials and technologies for their preparation.

Conclusion

A theoretical justification of the methodological approach for estimating the dimensional parameter determining the nanostate of a material object is proposed in the form of an analytical expression $L_0 = 230 \theta_D^{-1/2}$, which is based on the parameter of Debye temperature θ_D . This parameter determines the conditions for using classical or quantum representations to characterize condensed matter objects.

The use of the obtained expression, in our opinion, is justified. It takes into account the characteristics of the substance and is based on the Debye temperature parameter, which is either tabulated or can be calculated on the basis of experiments. The experimental and calculated data on the estimation of nanoscale parameters of dispersed particles are in satisfactory agreement. This indicates the validity of practical application of the proposed

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expression for selection of components of nanocomposite materials based on metal, polymer, ceramic and other matrices, and technologies for their

preparation to ensure the specified parameters of operational characteristics of products for various functional purposes.

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