MOLECULAR-STATISTICAL DESCRIPTION OF NONUNIFORMLY DEFORMED SPECIMENS. 2. CALCULATION OF THE DISTRIBUTION FUNCTIONS OF MOLECULES AND VACANCIES IN A ONE-DIMENSIONAL UNIFORMLY DEFORMED STATISTICAL EXTENSION-COMPRESSION MODEL

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Within the framework of a two-level molecular-statistical study of the thermodynamic and mechanical properties of condensed systems, a one-dimensional statistical model of uniform extension and compression of a crystal with vacancies has been developed. The micro- and macrostructures of the model are described using correlative distribution functions of real molecules (particles of the r (real) type) and vacancies, account of which is carried out using a subsystem of fictitious particles (quasiparticles of the f (fictitious) type) that do not interact with the molecules and with each other. A nonlinear integral equation for the average-force potentials which determine the single- and two-particle correlative functions of the two-component statistical system of real and fictitious particles has been obtained. The analytical solution of the integral equation has been found within the framework of a modified approach due to the vacancies of the Gauss approximation.

In recent times, it has become possible to construct a statistical theory of deformed crystal lattices with defects [1, 2] to which (in studying a microstructure) one cannot, in principle, apply the traditional methods of elasticity theory, whose foundation is formed by the concept of a continuous, i.e., model, medium. Therefore, the application of this theory is confined to a region of low deformations beyond which we have the unsolved problem of description of the nonlinear deformation of real bodies (with defects) with allowance for their plasticity and the actual absence of continuity of the material. If we consider the construction of a method to describe deformed real bodies (in crystalline and amorphous states) as our ultimate goal, we must solve a number of initial problems of fundamental character:

1) taking account of the discreteness of a material and the ensuing absence of continuity, at least at the lowest microscopic level of a theoretical description;

2) taking account of the structural change of a deformable medium with increase in the deformation and accumulation of defects of different types and the ensuing "production" of the entropy of a body;

3) calculation of the free energy of a deformed body which would take into account the structural features of a material (entropy factor) in addition to the potential strain energy (force factor).

Since the problems formulated belong to the domain of statistical physics, to solve them one must use modern statistical approaches, such as the method of correlative functions of Bogolyubov, Born, Green, Kirkwood, and Yvon (BBGKY) [3], L. A. Rott's method of conditional distributions [4], and the variational method of thermodynamic potentials [5], whose simultaneous employment has enabled us to develop a two-level molecular-statistical description of inhomogeneous systems, including crystals with defects [7, 8].

For practical implementation of the possibilities of the two-level molecular-statistical description of crystal specimens with vacancies it is desirable to have such an exactly solvable statistical model whose properties can be investigated by analytical methods. Since the state of a deformed crystal with vacancies is described, in the general case, by a rather complex nonlinear integral equation with field parameters (the field of microscopic deformation tensor and

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Fig. 1. Layout of nodes with occupied (solid circles) and vacant (open circles) nodes. $R = R_0(1 + \lambda)$.

the field of concentration of vacant lattice nodes) [6–8], to solve the problem formulated we restrict ourselves here to consideration of uniformly deformed linear specimens subjected to uniaxial extension and compression.

Description of the One-Dimensional Statistical Model of Uniaxial Uniform Deformation. For statistical modeling of uniform uniaxial extension and compression we consider a linear chain (Fig. 1) containing *M* microcells (of volume $\omega_l = \omega$, l = 1, 2, ..., M) in which *N* molecules are distributed. Since N < M, the subsystem of vacant cells (vacancies) in the statistical scheme developed enables us to rely on description of the contributions from all kinds of extended defects in real crystals that are formed due to the "condensation" of the vacancies under the action of internal force fields.

In the state of uniform extension with a relative deformation of $\lambda_l = \lambda$ for the molecular concentration $n_l^r = n_r \equiv n \ (n = N/M)$ (Fig. 1), the system of equations (9), in the approximation of interaction of only the nearest neighbors, is transformed to the following form (x and x' are the coordinates of the molecules reckoned from the centers of their cells of an undeformed lattice with a parameter R_0):

$$n \exp\left\{-\phi\left(x\right)\right\} \cong n_{\mathrm{rr}} \frac{\int_{\omega_{m}}^{\omega_{m}} \exp\left\{-\phi\left(x'\right)\right\} dx'}{\int_{\omega_{m}}^{\omega_{m}} \exp\left\{-\phi\left(x'\right)\right\} dx'} + n_{\mathrm{rf}} \frac{\int_{\omega_{l}}^{\omega_{l}} \exp\left\{-\phi\left(-x\right)\right\} dx}{\int_{\omega_{l}}^{\omega_{m}} \exp\left\{-\phi\left(-x\right)\right\} dx}.$$
(1)

To shorten the representation in (1) and in the subsequent relations we employ the overdetermined potentials of the average forces (φ) and intermolecular interaction (Φ) involving the temperature θ ; the potential φ depends on the deformation λ and the concentration *n*, while the potential Φ explicitly depends on the deformation λ as on the parameter

$$\varphi(x) \equiv \frac{1}{\theta} \varphi_{lm}(q_l, n, \lambda), \quad \Phi(\tilde{r}) = \frac{1}{\theta} \Phi(\tilde{r}_{lm}, \lambda) = \frac{4}{\theta} \left(\frac{1}{\tilde{r}^{12}} - \frac{1}{\tilde{r}^6} \right).$$
(2)

Here the mutual distance \tilde{r} for two molecules located near the neighboring nodes (centers of the cells) of a deformed chain (this is indicated by a ~ sign above *r*) is expressed in terms of the deformation λ and the coordinates *x* and x' of the molecules:

$$\tilde{r} = (R_0 + x - x')(1 + \lambda), \quad \lambda = \Delta R / R_0 = (R - R_0) / R_0.$$

It should be noted that the distances and the temperature are calculated, as is usually done, in the units of the parameters of the Lennard-Jones potential (σ is the length parameter and ε is the energy parameter, i.e., the potential-well depth for $\Phi(r)$, so that $\theta = kT/\varepsilon$).

Since we consider a uniform deformation of the chain, the general functional expression for two-cell occupation numbers is simplified (see formula (56) in [9]) for all pairs of neighboring cells:

$$n_{\rm rf} = \frac{1}{2z} \left(-1 + \sqrt{1 + 4n_{\rm r}n_{\rm f}z} \right), \quad z \equiv \langle \exp\left\{-\phi^{\rm r}\left(x\right)\right\} \rangle_l - 1 = \langle \exp\left\{-\phi\left(x\right)\right\} \rangle_l - 1 , \tag{3}$$

where

$$\exp\left\{-\phi^{r}(x)\right\} = \frac{\omega}{\int \exp\left\{-\phi(x')\right\} dx'} \int_{\omega} \exp\left\{-\phi(-x')\right\} dx'}.$$
(4)

Since the averaging in (1) and hence (3) and (4) is carried out using the auxiliary (see formula (10) from [1]) single-particle function $\tilde{F}_{11}^*(x)$, the quantity z is the average value of the Mayer function which is widely used in the nonideal-gas theory.

Let us apply the method of successive approximations and take into account that the concentration of the vacancies $n_{\rm f}$ in the crystal volume is low ($n_{\rm f} = 1 - n_{\rm r} < 10^{-3}$ at a large distance from the fusion line); therefore, we expand (3) in a series and restrict ourselves to its first terms in y = 4n(1 - n)z:

$$n_{\rm rf} \cong n \, (1-n) \, [1-n \, (1-n) \, z] \,. \tag{5}$$

In employing the iteration method for solution of the integral equation (1), as the first step we take into account that $n_{\rm rr} = n - n_{\rm rf} >> n_{\rm rf}$; therefore, we can disregard the second term on the right-hand side of (1) and rewrite (1) in a form which corresponds to the first iteration with the trial function $\varphi_0(x)$ substituted into the right-hand side of the integral equation. Then for the average-forces potential $\varphi_1(x)$ we obtain

$$\exp\left\{-\phi_{1}(x)\right\} \cong \frac{n_{\rm rr}}{n} \int_{-R_{0}/2}^{R_{0}/2} \exp\left\{-\phi_{0}(x')\right\} dx' \bigwedge_{-R_{0}/2}^{R_{0}/2} \exp\left\{-\phi_{0}(x')\right\} dx', \quad n_{\rm rr} = n - n_{\rm rf}.$$
(6)

Equation (6) still remains rather complex since it is strongly nonlinear due to the potential $\Phi(\tilde{r})$; moreover, the sought potential $\phi_1(x)$ depends on the concentration *n* and the relative deformation λ on which the mutual distance \tilde{r} for two molecules near the neighboring nodes of the deformed chain of *M* microcells depends explicitly.

Analytical Investigations of the One-Dimensional Extension–Compression Model in the Gauss Approximation. Taking into account that, in developing a new scientific trend, it is always desirable to have an exactly solvable model problem which allows its analytical investigation with subsequent refinement and generalization of the results, we expand the potentials $\varphi_0(x')$, $\varphi_1(x')$, and $\Phi(\tilde{r})$ as a series in the deviations x and x' of the molecules from the centers of their cells of an undeformed crystal lattice and restrict ourselves to the first three terms of the expansions; this corresponds to a modified Gauss approximation for correlative molecular-distribution functions in the statistical approach which takes into account the presence of the subsystem of vacancies (quasiparticles):

$$\phi_{0}(x') \approx \phi_{0} + \alpha_{0}x' + \beta_{0}x'^{2}, \quad \phi_{1}(x) \approx \phi_{1} + \alpha_{1}x + \beta_{1}x^{2},$$

$$\Phi(\tilde{r}) \approx \Phi_{0} + a(x - x') + b(x - x')^{2},$$
(7)

where the coefficients of expansion of the Lennard-Jones potential which depend on the deformation λ have the form

$$\Phi_{0} = \Phi(R) = \frac{4}{\theta} \left(\frac{1}{R_{0}^{12} (1+\lambda)^{12}} - \frac{1}{R_{0}^{6} (1+\lambda)^{6}} \right),$$

$$a = \frac{d\Phi(R)}{dR} (1+\lambda) = -\frac{24}{\theta} \left(\frac{2}{R_{0}^{13} (1+\lambda)^{12}} - \frac{1}{R_{0}^{7} (1+\lambda)^{6}} \right),$$

$$b = \frac{d^{2}\Phi(R)}{dR^{2}} \frac{(1+\lambda)^{2}}{2} = \frac{12}{\theta} \left(\frac{26}{R_{0}^{14} (1+\lambda)^{12}} - \frac{7}{R_{0}^{8} (1+\lambda)^{6}} \right).$$
(8)

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The performed expansion of the potentials (after their substitution into the integral equation (1)) enables us, in evaluating the integrals, to use the known reference tabular integral [10]

$$\int_{-\infty}^{\infty} \exp\left\{-\left(sx + px^{2}\right)\right\} dx = \sqrt{\frac{\pi}{p}} \exp\left\{\frac{s^{2}}{4p}\right\}, \quad p > 0.$$
(9)

To do this we must replace integration within the limits of each cell by integration within infinite limits; this is allowable since, as is known, the distribution functions of molecules in the crystalline state are strongly localized near the lattice nodes so that the standard (root-mean-square) deviation amounts to about 10% of the lattice parameter, i.e., $\sqrt{\langle x^2 \rangle} \sim 0.1R_0$. After such an evaluation of the integral on the right-hand side of (6) and equating the coefficients of x of the same power, we obtain the following system of equations for the sought coefficients of expansion of the potential φ_1 :

$$\varphi_1 = \Phi_0 + \frac{\alpha_0^2}{4\beta_0} - \frac{(\alpha_0 - a)^2}{4(b + \beta_0)} - \ln\left(\frac{n_{\rm rr}}{n}\sqrt{\frac{\beta_0}{b + \beta_0}}\right), \quad \alpha_1 = a + \frac{b(\alpha_0 - a)}{(b + \beta_0)}, \quad \beta_1 = b - \frac{b^2}{(b + \beta_0)}, \quad (10)$$

where $n_{\rm rr} = n - n_{\rm rf} \approx n^2 [1 + (1 - n)^2 z].$

The function \hat{F}_{11} of distribution of a molecule near the node and the auxiliary function \hat{F}_{11}^* in the first approximation have the following form:

$$\hat{F}_{11}(x) = n \frac{\exp\left\{-\left(\phi_{1} + \alpha_{1}x + \beta_{1}x^{2}\right)\right\} \exp\left\{-\left(\phi_{1} - \alpha_{1}x + \beta_{1}x^{2}\right)\right\}}{Q} = n \sqrt{\frac{2\beta_{1}}{\pi}} \exp\left\{-2\beta_{1}x^{2}\right\},$$

$$\hat{F}_{11}^{*}(x) = \frac{\exp\left\{-\left(\phi_{1} + \alpha_{1}x + \beta_{1}x^{2}\right)\right\}}{\int_{\infty}^{\infty} \exp\left\{-\left(\phi_{1} + \alpha_{1}x + \beta_{1}x^{2}\right)\right\} dx} = \sqrt{\frac{\beta_{1}}{\pi}} \exp\left\{-\left(\frac{\alpha_{1}^{2}}{4\beta_{1}}\right)\right\} \exp\left\{-\left(\alpha_{1}x + \beta_{1}x^{2}\right)\right\}.$$
(11)

Calculation of the Single-Particle Correlative Functions of the One-Dimensional Statistical Extension– Compression Model. Let us use a general functional statistical expression [6, 11] for the free energy of an inhomogeneous multicomponent system of molecules which contains a subsystem of vacancies. We apply it to the developed statistical model of uniaxial tension and compression [1] and evaluate the corresponding integrals in the quadratic approximation by the deviation x of the molecules from the nodes (centers of microcells). We obtain the following expression for the free energy of a deformed chain in the first approximation of a small parameter whose part is played by the second term in Eq. (1):

$$F \simeq \Theta M \left[n \ln n + (1 - n) \ln (1 - n) + n^{2} (1 - n)^{2} z^{2} \right] + \Theta M \left[\frac{n}{2} \left(\ln \left(\frac{\beta_{1}}{2\pi} \right) - \frac{\alpha_{1}^{2}}{\beta_{1}} \right) + \frac{n^{2}}{2} (1 + (1 - n)^{2} z) \left(\ln 2 + 2\varphi_{1} + \frac{\alpha_{1}^{2}}{2\beta_{1}} \right) \right].$$
(12)

The system of three equations (10) establishes a relationship between the six coefficients of the trial potential $\varphi_1(x)$ and the potential $\varphi_1(x)$ (φ_0 , α_0 , β_0 , φ_1 , α_1 , and β_1) which are found in solving the nonlinear integral equation by the iteration method.

The coefficient φ_0 is determined from the condition of normalization of the auxiliary function \hat{F}_{11}^* and does not appear in the final equations; therefore, the system contains only five unknown coefficients. Consequently, we must obtain two more equations of the relationship for the expansion coefficients. Let us use the available possibility



Fig. 2. Coefficients of expansion of the Lennard-Jones potential Φ_0 , *a*, and *b* (A) and the average-force potential β , β_1 , and ϕ_1 (B) vs. deformation λ (at $R_0 = 1$).

of relative freedom in selecting the trial function $\varphi_0(x)$ in the process of organization of an iteration procedure. We take into account that a computer-aided iteration procedure is completed if the sought function φ_1 coincides, with a sufficient degree of accuracy, with the previous function φ_0 employed on the right-hand side of the integral equation solved. Therefore, it seemed tempting to set $\varphi_1(x) = \varphi_0(x)$ and hence to equate the coefficients α and β in pairs, i.e., set $\alpha_1 = \alpha_0$ and $\beta_1 = \beta_0$, already on the first iteration of solution of the integral equation in the Gauss approximation. The analysis of such a possibility showed that $\beta_1 \neq \beta_0$ in the approximation employed, and we decided to determine the lacking equations of the relationship using the variational method which is based on the extremum properties of thermodynamic potentials of equilibrium systems, for example, free energy.

From expression (12) it is seen that it contains the term *n*, which is linear in concentrations, in the second square brackets; this term must, probably, be absent in a rigorous expression for free energy, since the contribution from pair interactions in the limit for a vanishing concentration *n* (when the correlation in occupation of the pairs of cells can be disregarded) must be quadratic in *n*. Therefore, by equating the coefficient of n/2 (in the parentheses) to zero, we obtain an additional equation of the relationship between the coefficients α_1 and β_1 :

$$\ln\left(\frac{\beta_1}{2\pi}\right) - \frac{\alpha_1^2}{\beta_1} = 0.$$
⁽¹³⁾

Since the second equation from system (10) has the solution for $\alpha_1 = \alpha_0 = a$, the system is simplified:

$$\varphi_1 = \Phi_0 + \frac{\alpha_0^2}{4\beta_0} - \ln\left(\frac{n_{\rm rr}}{n}\sqrt{\frac{\beta_0}{b+\beta_0}}\right),\tag{14}$$

$$\beta_1 = b - b^2 / (b + \beta_0) , \qquad (15)$$

where

$$n_{\rm rr} \approx n^2 \left[1 + (1-n)^2 z\right], \quad z = \sqrt{\frac{1}{2}} \exp\left\{-\left(\varphi_1 + \frac{\alpha_1^2}{4\beta_1^2}\right)\right\} - 1,$$

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Fig. 3. Normalized unary function $F_{11}(x)$ (A) and the auxiliary function $F_{11}(x)$ (B) showing the distribution of a particle in the vicinity of the microcell node with account for the fact that the neighboring cell is vacant for different deformations λ and temperature θ .

and, together with Eq. (13), forms a complete system of three equations for the coefficients ϕ_1 , β_1 , and β_0 . The quantities Φ_0 , *a*, and *b* appearing in Eqs. (14) and (15) are the coefficients of expansion of the Lennard-Jones potential, which involves the temperature (see formula (2)) (Fig. 2). Therefore, they are known functions of the deformation λ and the temperature θ .

We have developed a block diagram and composed a computer program with the use of the Mathematica 3.0 application package to solve a system of nonlinear equations. System (13)–(15) was solved in the following order:

(1) the values of the reduced temperature θ (in the units of the parameter ε), the concentration *n*, and the deformation λ (for $R_0 = 1$ in the units of the parameter σ) were prescribed;

(2) the coefficients Φ_0 , *a*, and *b* were calculated from formulas (8);

(3) using the Mathematica 3.0 package the transcendental equation (13) was solved relative to the coefficient β in the approximation $\alpha \cong a$ and then the coefficient β_0 was determined from Eq. (15);

(4) the nonlinear transcendental equation (14) was solved relative to the coefficient φ_1 with the use of the Mathematica 3.0 package as well;

(5) the values found for all the coefficients were employed to calculate the distribution functions and the twocell numbers of occupation of the pairs of neighboring cells.

The results of the analogous calculations for other values of the deformation λ enabled us to establish the dependence of all the coefficients on the relative deformation λ (at the prescribed values of the temperature θ and the concentration *n*). These dependences are graphically presented in Fig. 2.

The solutions obtained make it possible to establish the influence of the temperature and the concentration of vacancies on the structure of the distribution of linear-chain particles. In the two-level approach employed, the chain structure is described using the normalized unary distribution functions $\hat{F}_{11}(x)$ and auxiliary distribution function $\hat{F}_{11}(x)$ (this function appears in the nonlinear integral equation for the average-force potential). In the quadratic ap-



Fig. 4. Correlator z and two-cell occupation numbers $n_{\rm rf}$ vs. deformation λ (for fixed temperature $\theta = 1$ and concentrations n: 1) $n_{\rm r} = 0.96$ and 2) 0.999).

proximation by displacements (Gauss approximation) for a linear chain these functions have the following form (see formulas (11)):

$$\hat{F}_{11}(x) = n \sqrt{\frac{2\beta_1}{\pi}} \exp\left\{-2\beta_1 x^2\right\},$$
(16)

$$\hat{F}_{11}^{*}(x) = \sqrt{\frac{\beta_1}{\pi}} \exp\left\{-\left(\alpha_1^2/4\beta_1\right)\right\} \exp\left\{-\left(\alpha_1 x + \beta_1 x^2\right)\right\}.$$
(17)

The analytical expression for function (16) with account for the dependence of the coefficient β_1 on the deformation λ (Fig. 2B) shows that as the deformation increases with extension of the linear chain the distribution function $F_{11}(x)$ becomes "smeared" (Fig. 3A) while the standard deviation of the molecule from the node increases, since

$$\langle x^2 \rangle = \int x^2 \hat{F}_{11}(x) \, dx = n \, \sqrt{\frac{2\beta_1}{\pi}} \, \frac{1}{2} \, \sqrt{\frac{\pi}{(2\beta_1)^3}} = \frac{n}{8\beta_1}.$$
 (18)

Function (17) describes the molecular distribution in a cell neighboring the vacant node of the chain. From (17) it is seen that it is asymmetric with respect to the node, i.e., the center of the cell (Fig. 3B). In this case, the dependence of the standard deviation on the deformation λ has a more complex form, since this deviation is expressed in terms of two coefficients now: α_1 ($\alpha_1 \cong a$) and β_1 ; the values of the coefficient β_1 and of the modulus of the coefficient α_1 decrease as λ increases (Fig. 2). Figure 4 shows the dependence of the correlator z and the two-cell occupation numbers $n_{\rm rf}$ on the deformation λ which determine the probability that there is a vacancy near the occupied cell. It is seen that as λ increases the quantity $n_{\rm rf}$ remains constant, in practice, for a fixed concentration n. However, it is clear that in the case of deformation the concentration of vacancies c = 1 - n changes, which will lead to the corresponding dependence of $n_{\rm rf}$ on λ . To establish the form of this dependence we are to solve the variational problem on determination of the minimum free energy of a deformed chain.

Thus, the above calculations show that the proposed one-dimensional statistical model of uniaxial extension and compression makes it possible to take into account, in explicit form, the influence of the interparticle interaction, the temperature, and the deformation on the microstructure and hence on the thermodynamic properties. Since the free energy depends on the deformation λ , this influence will also have an effect on the mechanical properties of a deformable specimen. This means that we have the conditions for construction of the foundations of a statistical elasticity theory that will be free of a model phenomenological idea of the continuity of a material (at a microlevel, a medium has the discreteness of a structure) and will be capable of describing nonlinear portions of an extension–compression diagram, which, as is known, are responsible for nonlinear elasticity.

NOTATION

N, number of particles of a given system; M, number of microcells into which the entire volume of the system is mentally subdivided; λ , relative deformation of the specimen; n, molecular concentration in the microcells; ω , volume of a microcell; ω_M , volume of the last microcell; φ , potential of the average forces of interaction of the particles in the microcells; Φ , pair intermolecular Lennard-Jones potential; $\theta = kT/\epsilon$, dimensionless temperature (k, Boltzmann constant, T, absolute temperature, ε , depth of the potential well that describes the interaction of two particles (molecules) of the medium); x, x', and q, coordinate scalars (positions) of a point of the medium relative to the center of the cell of the undeformed lattice of the specimen; R, parameter of the deformed lattice; R_0 , parameter of the undeformed lattice; r, mutual distance for two molecules located near the neighboring nodes (centers of the cells) of the specimen; z, average value of the Mayer function; s and p, parameters of the tabular integral; F_{11} , conditional correlative distribution functions of molecules near the cell node; φ_0 , α_0 , and β_0 , coefficients of expansion of interaction of mean forces $\phi_0(x')$; ϕ_1 , α_1 , and β_1 , same, $\phi_1(x)$; Φ_0 , a, and b, coefficients of expansion of the Lennard-Jones potential; F, free energy of the deformed chain. Subscripts and superscripts: r, microcell occupied by a particle (real particle); f, (vacant) cell occupied by a fictitious particle; l and m, numbers of microcells; \sim , membership of the corresponding quantities in the deformed specimen; lm, pairs of cells; rr, two microcells with the numbers l and m are filled with one particle (molecule); rf, microcell with the number l is filled with one particle, while the microcell with the number m is empty (vacant); 11, first statistical approximation in which all microcells are occupied by no more than one particle (molecule); $^{\wedge}$, normalization to unity; * , auxiliary nature.

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