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Thermodynamics of the Fermi Gas in a Sphere of Arbitrary Radius

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For the Fermi gas filling the space inside the spherical cavity of arbitrary radius and with an arbitrary, including small, number of particles, the thermodynamic characteristics are calculated in the general form, namely: entropy, energy, thermodynamic potential, constituent elements of equation of state, heat capacities, and compressibilities. The size effects relating to the discrete structure of levels are investigated. On the example of the two- and three-level models, there is traced both the sequence of level filling with increasing temperature and the temperature dependence of the chemical potential. As shown, at temperatures, at which the filling of a new level begins, the heat capacity undergoes jumps.

Key words: Fermi particles, low-dimensional system, thermodynamic functions, entropy, equation of state, heat capacity, compressibility, discrete levels.

Для Фермі-газу, що заповнює простір усередині сферичної порожнини довільного радіюса та за довільного, зокрема малого, числа частинок, у загальному вигляді обчислено термодинамічні характеристики, а саме: ентропію, енергію, термодинамічний потенціял, складові рівняння стану, тепломісткості та стисливості. Досліджено розмірні ефекти, пов'язані з дискретною структурою рівнів. На прикладі дво- та трирівневого моделів простежено послідовність заповнення рівнів зі зростанням температури та температурну залежність хемічного потенціялу. Показано, що за температур, за яких починається заповнення нового рівня, мають місце стри-

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бки тепломісткости.

Ключові слова: Фермі-частинки, низькорозмірна система, термодинамічні функції, ентропія, рівняння стану, тепломісткість, стисливість, дискретні рівні.

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1. INTRODUCTION

The ideal Fermi gas model is the basis for understanding the properties of metals, electron and other multifermion systems. In many cases, it is also possible to describe with acceptable accuracy the behaviour of systems of interacting Fermi particles within the framework of the approximation of an ideal gas of quasiparticles, whose dispersion law differs from the dispersion law of free particles. It is essential that all thermodynamic characteristics of the ideal Fermi gas at arbitrary temperatures in the case of a large volume can be expressed through special functions and, thus, all relations of phenomenological thermodynamics can be obtained and checked within the framework of the quantum microscopic model.

So far, increasing attention is paid to the study of quantum properties of systems with a small number of particles, such as quantum dots, nanostructured and mesoscopic objects. In this connection, the problem of description of such small objects with taking into account their interaction with the external environment becomes actual. Statistical description is usually used to study systems with a very large number of particles. However, statistical methods can also be applied in the study of equilibrium states of systems with a small number of particles and even a single particle. When considering a system within a large canonical ensemble, it is assumed that it is a part of a very large system, a thermostat, with which it can exchange energy and particles. The thermostat itself is characterized by such statistical quantities as temperature T and chemical potential μ. Assuming that the subsystem under consideration is in thermodynamic equilibrium with the thermostat, the subsystem itself, even consisting of a small number of particles, will be characterized by the same quantities. For example, one can consider the thermodynamics of a single quantum oscillator [1]. In the case, when an exchange of particles with the thermostat is possible, the time-averaged number of particles of a small subsystem may be non-integer and, in particular, even less than unity.

In statistical physics, the entropy and distribution functions of particles over quantum states are calculated under the assumption that the number of particles is very large. Such consideration for fermions leads to the Fermi–Dirac distribution, and for bosons, to the Bose–Einstein distribution [1]. In work [2], the authors calculated the entropy and distribution functions of non-interacting particles in the case when no restrictions are imposed on their number in a system being in thermodynamic equilibrium with the environment. In Ref. [3], there were studied in detail the thermodynamic properties of a two-level system of finite volume, and in Refs. [4, 5], those properties of the Fermi gas at arbitrary temperatures and number of particles filling the space inside the cubic cavity of fixed volume were studied too. The thermodynamic characteristics were calculated, the discrete structure of energy levels was taken into account and size effects at low temperatures were studied.

The thermodynamic properties of large three-dimensional systems do not depend on the shape of the occupied volume. On the contrary, in systems of small size where the discrete structure of the spectrum is essential, the thermodynamic characteristics also depend on the shape of the occupied volume. This work, in development of work [5], is devoted to the study of the thermodynamic characteristics of the Fermi gas filling a spherical cavity of an arbitrary radius and with an arbitrary, including small, number of particles. The thermodynamic characteristics such as entropy, energy, thermodynamic potential, equation of state, heat capacities and compressibilities are calculated in the general form. The size effects, connected with the discrete structure of levels and leading to the heat capacity jumps, are investigated.

2. SCHRÖDINGER EQUATION FOR THE SPHERE

Before considering a multiparticle system, we present the solution of the Schrödinger equation for a single particle in a sphere of radius a. No restrictions are imposed on the radius of the sphere, and, therefore, the approach used below is applicable to the study of objects of small size. In spherical coordinates, a particular solution of the Schrödinger equation for the state characterized by the orbital momentum l, magnetic number m and wave number k has the form $\psi_{klm}(r,\Omega) = Bj_l(kr)Y_{lm}(\Omega)$ [6], where $Y_{lm}(\Omega)$ are the spherical functions, $\Omega \equiv (\varphi,\theta)$, and $j_l(kr) = \sqrt{\pi/2kr}\,J_{l+1/2}(kr)$ is the spherical Bessel function satisfying the equation

$$\frac{d^2 j_l}{dr^2} + \frac{2}{r}\frac{dj_l}{dr} + \left(k^2 - \frac{l(l+1)}{r^2}\right)j_l = 0\,. \tag{1}$$

It is assumed that the spin of the Fermi particle is equal to S, and the potential barrier on the surface of the sphere is infinite, so that the wave function of the particle at the boundaries turns to zero. Taking into account the boundary condition $\psi_{klm}(a,\Omega) = 0$ or $j_l(ka) = 0$, we find that the wave number can take an infinite number of discrete values of $k_{l\alpha} = g_{l\alpha}/a$, and the energy of the particle in such a state is determined by the formulae

$$\varepsilon_{l\alpha} \equiv \varepsilon_0 g_{l\alpha}^2, \; \varepsilon_0 = \frac{\hbar^2}{2ma^2}. \tag{2}$$

The non-zero values of the numbers $g_{l\alpha}$, at which the Bessel function turns to zero, are described by the orbital momentum l and the index $\alpha = 1, 2, \ldots$ numbering the zeros in the ascending order. The following inequalities hold for the zeros:

$$\begin{gathered} l < g_{l1} < g_{l2} < g_{l3} < \ldots, \; g_{l\alpha_1} < g_{l\alpha_2} \; (\alpha_1 < \alpha_2), \\ g_{l1} < g_{l+1,1} < g_{l2} < g_{l+1,2} < g_{l3} < g_{l+1,3} < \ldots \; (g_{l\alpha} < g_{l+1,\alpha}). \end{gathered} \tag{3}$$

For brevity, a zero with given l and α will sometimes be denoted by the single index $j \equiv (l, \alpha)$.

The smallest zero is $g_{\min} \equiv g_{01} = \pi$. The energy levels are doubly degenerate in the spin projection and $(2l + 1)$-multiply degenerate in the magnetic quantum number, so that the total degeneracy factor of the level $j \equiv (l, \alpha)$ is equal to $z_j = 2(2l + 1)$. The first ten zeros of the Bessel function and the degeneracy factors of the corresponding levels are given in Table 1.

To obtain an approximate formula for the zeros at large α, one should use the asymptotics $j_l(x) \approx x^{-1} \sin(x - l\pi/2)$, which gives

$$g_{l\alpha} = \frac{\pi}{2}(l + 2\alpha). \tag{4}$$

The normalization factor B of the wave function is found from condition $B^2 \int_0^a dr r^2 j_l^2(kr) = 1$; so, such a wave function takes the form

$$\psi_{lm\alpha}(r, \Omega) \equiv f_{l\alpha}(r) Y_{lm}(\Omega), \; f_{l\alpha}(r) \equiv \frac{\sqrt{2}}{a^{3/2} Z_l^{1/2}(g_{l\alpha})} j_l\left(g_{l\alpha}\frac{r}{a}\right), \tag{5}$$

TABLE 1. The bottom ten energy states inside the sphere.

j	1	2	3	4	5	6	7	8	9	10
(l,α)	(0,1)	(1,1)	(2,1)	(0,2)	(3,1)	(1,2)	(4,1)	(2,2)	(5,1)	(0,3)
g_j	3.142	4.493	5.763	6.283	6.988	7.725	8.183	9.095	9.356	9.425
g_j^2	9.872	20.19	33.21	39.48	48.83	59.68	66.96	82.72	87.54	88.83
z_j	2	6	10	2	14	6	18	10	22	2

where $Z_l(g_{l\alpha}) = -j_{l+1}(g_{l\alpha})\, j_{l-1}(g_{l\alpha})$.

3. DISTRIBUTION FUNCTION FOR AN ARBITRARY NUMBER OF PARTICLES

The equations for the populations of levels in the Fermi gas for an arbitrary, even small, number of particles were obtained by the authors in Refs. [2, 3, 5]. When constructing the thermodynamics of a system with an arbitrary number of particles and an arbitrary size, we proceed from the combinatorial expression for entropy. If, at each level of the quantum Fermi system with energy ε_j and degeneracy factor z_j, there are N_j particles, then, the entropy is determined by the formula [1]

$$S = \sum_j \left[\ln z_j! - \ln N_j! - \ln\left(z_j - N_j\right)! \right]. \tag{6}$$

To calculate all factorials at $N >> 1$, it is usually applied the Stirling's formula in the form $\ln N! \approx N \ln(N/e)$. When N is small, the accuracy of this formula is not sufficient. In case, when, as it is supposed, the time-averaged number of particles can be arbitrary, in particular, small and fractional, the factorial should be defined through the gamma function [7]:

$$N! = \Gamma(N+1). \tag{7}$$

From here, it follows the formula for the nonequilibrium entropy:

$$S = \sum_j S_j\,,\; S_j = -\ln\Gamma\left(z_j n_j + 1\right) - \ln\Gamma\left[z_j\left(1 - n_j\right) + 1\right] + \ln\Gamma\left(z_j + 1\right). \tag{8}$$

The equation that determines the average number of particles in each state contributing to the entropy [2, 3, 5] is found from the requirement of extremum of the entropy under the condition of constancy of the number of particles and energy

$$\theta\left(n_j, z_j\right) = \frac{\left(\varepsilon_j - \mu\right)}{T}, \tag{9}$$

where

$$\theta\left(n_j, z_j\right) \equiv \theta_j \equiv \psi\left[z_j\left(1 - n_j\right) + 1\right] - \psi\left(z_j n_j + 1\right), \tag{10}$$

T is a temperature, μ is a chemical potential, $\psi(x) = d\ln\Gamma(x)/dx$ is the logarithmic derivative of the gamma function (the psi function) [7]. If a level is filled ($n_j = 1$) or empty ($n_j = 0$), then, it does not contribute to the total entropy, and therefore, Eq. (9) applies only to partially filled levels. It is also important to note that the exact function n_j determined by Eq. (9) tends to zero or unity at finite values of energy.

4. THERMODYNAMIC FUNCTIONS

The total number of particles N, which, as noted, is considered as a continuous positive quantity taking also fractional values, and the total energy E are determined by the formulae

$$N = \sum_j N_j = \sum_j n_j z_j \,, \tag{11}$$

$$E = \sum_j \varepsilon_j N_j = \sum_j \varepsilon_j n_j z_j \,. \tag{12}$$

The average number of particles $n_j = N_j/z_j$ at level j, or the population of the level, is found from Eqs. (9). When solving this set of equations, one should take into account the condition of constancy of the total number of particles (11). The differential of the thermodynamic potential $\Omega = E - TS - \mu N$ has the usual form

$$d\Omega = -SdT - Nd\mu - pdV \,, \tag{13}$$

where the pressure

$$p = -\sum_j z_j n_j \frac{d\varepsilon_j}{dV} \tag{14}$$

is determined by the dependence of the particle energy on the volume $V = 4\pi a^3/3$. In the given case, $d\varepsilon_i/dV = -2\varepsilon_i/3V$; so, $p = 2E/3V$. To calculate heat capacities and thermodynamic coefficients, we first find the differentials of the distribution function, number of particles, entropy and pressure:

$$z_j dn_j = \frac{\theta_j}{\theta_j^{(1)}}\frac{dT}{T} - \frac{1}{\theta_j^{(1)}}\frac{d\varepsilon_j}{dV}\frac{dV}{T} + \frac{1}{\theta_j^{(1)}}\frac{d\mu}{T} \,, \tag{15}$$

$$dN = \frac{dT}{T}\sum_j \frac{\theta_j}{\theta_j^{(1)}} - \frac{dV}{T}\sum_j \frac{1}{\theta_j^{(1)}}\frac{d\varepsilon_j}{dV} + \frac{d\mu}{T}\sum_j \frac{1}{\theta_j^{(1)}} \,, \tag{16}$$

$$dS = \frac{dT}{T}\sum_j \frac{\theta_j^2}{\theta_j^{(1)}} - \frac{dV}{T}\sum_j \frac{\theta_j}{\theta_j^{(1)}}\frac{d\varepsilon_j}{dV} + \frac{d\mu}{T}\sum_j \frac{\theta_j}{\theta_j^{(1)}} \,, \tag{17}$$

$$dp = -\frac{dT}{T}\sum_j \frac{\theta_j}{\theta_j^{(1)}}\frac{d\varepsilon_j}{dV} + \frac{dV}{T}\sum_j \left[\frac{1}{\theta_j^{(1)}}\left(\frac{d\varepsilon_j}{dV}\right)^2 - z_j n_j T \frac{d^2\varepsilon_j}{dV^2}\right] - \frac{d\mu}{T}\sum_j \frac{1}{\theta_j^{(1)}}\frac{d\varepsilon_j}{dV} \,; \tag{18}$$

$$\theta_j^{(1)} \equiv \theta^{(1)}\left(n_j, z_j\right) \equiv \psi^{(1)}\left[z_j\left(1 - n_j\right) + 1\right] + \psi^{(1)}\left(z_j n_j + 1\right), \tag{19}$$

where $\psi^{(1)}(y) = d\psi(y)/dy = d^2 \ln\Gamma(y)/dy^2$ is the trigamma function [7].

In the following, we will consider systems with a fixed average number of particles, for which $dN = 0$. This condition allows eliminating the differential of chemical potential and, as a result, the entropy and pressure differentials will take the form

$$dS = B_T \frac{dT}{T} + B_V \frac{dV}{T}, \quad dp = A_T \frac{dT}{T} + A_V \frac{dV}{T}, \tag{20}$$

where

$$B_T = \sum_j \frac{\theta_j^2}{\theta_j^{(1)}} - \theta^{(1)} \left(\sum_j \frac{\theta_j}{\theta_j^{(1)}} \right)^2, \quad B_V = A_T = \theta^{(1)} \left(\sum_j \frac{\theta_j}{\theta_j^{(1)}} \right) \left(\sum_j \frac{1}{\theta_j^{(1)}} \frac{d\varepsilon_j}{dV} \right) - \left(\sum_j \frac{\theta_j}{\theta_j^{(1)}} \frac{d\varepsilon_j}{dV} \right),$$

$$A_V = -T \sum_j z_j n_j \frac{d^2\varepsilon_j}{dV^2} - \theta^{(1)} \left(\sum_j \frac{1}{\theta_j^{(1)}} \frac{d\varepsilon_j}{dV} \right)^2 + \sum_j \frac{1}{\theta_j^{(1)}} \left(\frac{d\varepsilon_j}{dV} \right)^2, \quad \frac{1}{\theta^{(1)}} \equiv \sum_j \frac{1}{\theta_j^{(1)}}. \tag{21}$$

In our case, $d\varepsilon_i/dV = -2\varepsilon_i/3V$, $d^2\varepsilon_i/dV^2 = 10\varepsilon_i/9V^2$. From these relations, there follow the formulae for the isochoric heat capacity,

$$C_V = T \left(\frac{\partial S}{\partial T} \right)_{N,V} = B_T, \tag{22}$$

and the isobaric one,

$$C_p = T \left(\frac{\partial S}{\partial T} \right)_{N,\,p} = B_T - \frac{B_V^2}{A_V}. \tag{23}$$

Let us also present, following from Eqs. (20), (21), formulae for the coefficient of volumetric expansion,

$$\alpha_p = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p = -\frac{B_V}{VA_V}, \tag{24}$$

the isothermal compressibility,

$$\gamma_T = -\frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_T = -\frac{T}{VA_V}, \tag{25}$$

and the isochoric thermal pressure coefficient,

$$\beta_V = \frac{1}{p} \left(\frac{\partial p}{\partial T} \right)_V = \frac{B_V}{pT}. \tag{26}$$

All other thermodynamic coefficients can be expressed [8] through the heat capacities and the coefficients given here. Obviously, the general relation

$$C_p - C_V = TV\frac{\alpha_p^2}{\gamma_T} = -\frac{B_V^2}{A_V} \tag{27}$$

holds [1] that confirms the consistency of the given general thermodynamic relations. Since the thermodynamic inequalities $C_V > 0$ and $\left(\partial p/\partial V\right)_T < 0$ must be satisfied in a stable system [1], then, there must be $B_T > 0$ and $A_V < 0$.

5. LOW TEMPERATURES. SIZE EFFECTS

Of most interest is the case, when there is a small number of particles in the volume of a sphere of small radius. In this case, it is important to take into account the discrete structure of the spectrum, so that the exact formulae (9), (14), (21)–(26) should be used in calculations. In the following, along with dimensional quantities, we will use the notation of quantities in dimensionless form, introducing arbitrary characteristic scales of radius a_*, energy $\varepsilon_{0*} = \hbar^2/2ma_*^2$ and pressure $p_* \equiv \varepsilon_{0*}/2\pi a_*^3$. Note that $a_*^2\varepsilon_{0*} = a^2\varepsilon_1/g_1^2$. The dimensionless radius a, temperature τ, pressure p and level energy $\tilde{\varepsilon}_j$ are defined by the relations:

$$\tilde{a} = \frac{a}{a_*},\ \tau = \tilde{a}^2\frac{T}{\varepsilon_{0*}},\ \tilde{p} = \frac{p}{p_*},\ \tilde{a}^2\frac{\varepsilon_j}{\varepsilon_{0*}} = g_j^2. \tag{28}$$

In these notations, Eqs. (9) will take the form

$$\theta\left(n_j, z_j\right) = \frac{g_j^2 - \tilde{\mu}}{\tau}, \tag{29}$$

where $\tilde{\mu} = \tilde{a}^2\mu/\varepsilon_{0*}$ is the dimensionless chemical potential.

Let us first consider the state of the system at zero temperature. From the energy minimum condition it follows that, if the number of particles is less than or equal to the degeneracy factor of the first level, $0 < N \le z_1$, then, $\mu = \varepsilon_1$, $0 < n_1 \le 1$, and the particles are only at the ground level. Higher levels are not occupied. In this case, the population of the first level n_1 is determined by the relation $N = z_1 n_1$, and the energy, pressure, and thermodynamic coefficients are equal to $E = \varepsilon_1 z_1 n_1$, $p = 2E/3V$, $\alpha_p = 0$, $\beta_V = 0$, $\gamma_T = (3/5)\,p^{-1}$. The entropy

$$S = -\ln\Gamma\left(z_1 n_1 + 1\right) - \ln\Gamma\left[z_1\left(1 - n_1\right) + 1\right] + \ln\Gamma\left(z_1 + 1\right) \tag{30}$$

turns to zero only for the fully occupied level ($n_1 = 1$) and is distinct from zero for the unfilled level. The dependences of the entropy on the population n for several values of the degeneracy factor z are shown in Fig. 1.

In this case, the third law of thermodynamics is satisfied in the Nernst formulation, while it is satisfied only at fully occupied levels in the Planck formulation.

If $M-1$ lower levels are completely filled and level M can be partially filled, then, $\sum_{j=1}^{M-1} z_j < N \le \sum_{j=1}^{M-1} z_j + z_M$ and $0 < n_M \le 1$, and the chemical potential $\mu = \varepsilon_M$. In this case, the entropy is given by Eq. (30) with the substitution $n_1 \to n_M$, and the total number of particles, energy, pressure, and thermodynamic coefficients are determined by formulae $N = \sum_{j=1}^{M-1} z_j + n_M z_M$, $E = \sum_{j=1}^{M-1} \varepsilon_j z_j + n_M \varepsilon_M z_M$, $p = 2E/3V$, $\gamma_T = 3/(5p)$, $\alpha_p = 0$, $\beta_V = 0$.

The discreteness of levels, which is a consequence of taking into account the finite size of the system, leads to the situation that near zero temperature there is a temperature region in which the populations of levels do not change with temperature and remain the same as at $T = 0$ K. With a further increase in temperature, there begin to occur transitions of particles to higher levels.

In the temperature region, where the temperature dependence of populations arises, the quantities (21), through which the heat capacities and thermodynamic coefficients (22)–(26) are expressed, can be represented in dimensionless variables in the form

$$B_T = \frac{G}{\tau^2}, \quad B_V = A_T = \frac{\varepsilon_{0*}}{2\pi\tau\tilde{a}^5 a_*^3} G, \quad A_V = \frac{\varepsilon_{0*}^2}{4\pi^2 \tilde{a}^{10} a_*^6}\left(G - \frac{5}{2}\tau d\right), \tag{31}$$

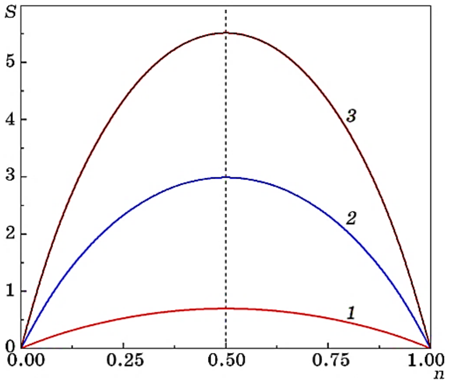

Fig. 1. Dependence of the entropy $S(n)$ on the level population at zero temperature for three values of the level degeneracy factor: *1*—$z = 2$; *2*—$z = 6$; *3*—$z = 10$.

where the following notations are used:

$$d \equiv \sum_j z_j g_j^2 n_j, \;\; G \equiv h_4 - \theta^{(1)} h_2^2,$$
$$h_2 \equiv \sum_j \frac{g_j^2}{\theta^{(1)}\left(n_j, z_{jj}\right)}, \;\; h_4 \equiv \sum_j \frac{g_j^4}{\theta^{(1)}\left(n_j, z_j\right)}, \;\; \frac{1}{\theta^{(1)}} \equiv \sum_j \frac{1}{\theta^{(1)}\left(n_j, z_j\right)}. \qquad (32)$$

Note that the quantity G does not explicitly contain the chemical potential and is positive due to the requirement of thermodynamic stability. The coefficient A_V must be negative ($A_V < 0$) for the thermodynamically stable system; this entails the fulfilment of the inequality $\tau > \tau_*$, where

$$\tau_* \equiv \frac{2}{5}\frac{G}{d}. \qquad (33)$$

At $\tau < \tau_*$, the excited system is unstable, and so the system continues to remain in the same ground state as at $T = 0$ K. Thus, the calculation of the temperature dependence of any thermodynamic quantity is reduced to the calculation of sums (32). In particular, the pressure (14) is

$$\frac{p}{p_*} = \frac{d}{\tilde{a}^5}. \qquad (34)$$

Let us consider the excitation of a system with a small number of particles at increasing temperature on the examples of simple two-level and three-level systems.

6. TWO-LEVEL SYSTEM

Thermodynamic properties of the two-level system in the general case were considered by the authors in work [3]. In the case of a spherical cavity, the degeneracy factors of the first two levels are $z_1 = 2,\ z_2 = 6$. As the temperature increases, the chemical potential first changes linearly with temperature, and the particles continue to remain at the same levels as at $T = 0$ K. Subsequently, transitions begin to occur from lower levels to higher ones. The temperature, at which there begins the transition from levels $i_1, i_2, \ldots, i_{m_i}$ to levels $j_1, j_2, \ldots, j_{m_j}$ at a total number of particles N, will be denoted as $\tau^{(N)}_{i_1 \ldots i_{m_i}, j_1 \ldots j_{m_j}}$, and the corresponding chemical potential will be denoted as $\tilde{\mu}^{(N)}_{i_1 \ldots i_{m_i}, j_1 \ldots j_{m_j}}$. The temperature and the corresponding chemical potential, at which levels $i_1, i_2, \ldots, i_{m_i}$ become empty, will be denoted as $\tau^{(N)}_{0 i_1 \ldots i_{m_i}}$ and $\tilde{\mu}^{(N)}_{0 i_1 \ldots i_{m_i}}$.

First, let us consider the case, when the number of particles is less than the degeneracy factor of the first level, so that the second level at $T = 0$ K is not filled: $N = z_1 n_1 \le z_1,\ n_2 = 0$. The transitions of particles from the first level to the second one will begin at the following values of temperature and chemical potential

$$\tau_{1,2}^{(N)} = \frac{g_2^2 - g_1^2}{\theta(0, z_2) - \theta(N/z_1, z_1)},\quad \tilde{\mu}_{1,2}^{(N)} = \frac{g_1^2\theta(0, z_2) - g_2^2\theta(N/z_1, z_1)}{\theta(0, z_2) - \theta(N/z_1, z_1)}. \tag{35}$$

Note that the functions (10) entering into formulae can be represented in the form

$$\theta(n, z) = \int_0^1 \frac{t^{zn} - t^{z(1-n)}}{1 - t}\, dt. \tag{36}$$

At $\tau > \tau_{1,2}^{(N)}$, the state is described by two equations $\tilde{\mu} = g_1^2 - \tau\theta(n_1, z_1) = g_2^2 - \tau\theta(n_2, z_2)$ with account of the constancy of the number of particles: $N = z_1 n_1 + z_2 n_2$. These equations allow finding the temperature dependences of the chemical potential $\tilde{\mu} = \tilde{\mu}(\tau)$ and the level populations $n_1 = n_1(\tau),\ n_2 = n_2(\tau)$, which, for the case $N \le z_1$, are shown in Figs. 2*a* and 3*a*, respectively.

If $N < N_c = 0.753$, then, at the values

$$\tau_{01}^{(N)} = \frac{g_2^2 - g_1^2}{\theta(N/z_2, z_2) - \theta(0, z_1)},\quad \tilde{\mu}_{01}^{(N)} = \frac{g_1^2\theta(N/z_2, z_2) - g_2^2\theta(0, z_1)}{\theta(N/z_2, z_2) - \theta(0, z_1)}, \tag{37}$$

all particles transit to the upper level, and the ground level becomes empty. If the above inequality is not satisfied, then, particles are present at both levels at all temperatures. The critical value of the particle number N_c is found from the equation $1/\tau_{01}^{(N_c)} = 0$.

In another case, when, at $T = 0$ K, the lower level is completely filled and the upper level is only partially filled, $N = z_1 + N',\ 0 < N' \le z_2,\ n_1 = 1,\ n_2 = N'/z_2$, then, the parameters, at which the transition from the first level to the second one begins, are determined by the formulae

$$\tau_{1,2}^{(N)} = \frac{g_2^2 - g_1^2}{\theta(N'/z_2, z_2) - \theta(1, z_1)},\quad \mu_{1,2}^{(N)} = \frac{g_1^2\theta(N'/z_2, z_2) - g_2^2\theta(1, z_1)}{\theta(N'/z_2, z_2) - \theta(1, z_1)}. \tag{38}$$

In this case, the lower level remains filled at all temperatures. The temperature dependences of the chemical potential and populations for this case are shown in Figs. 2*b* and 3*b*, respectively.

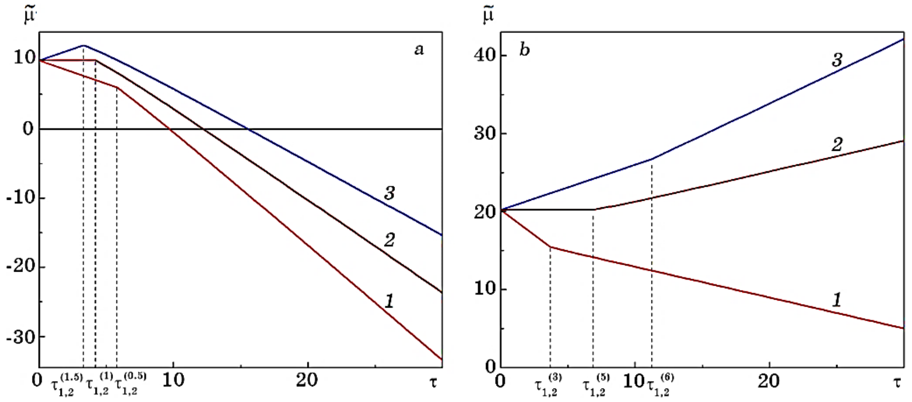

Fig. 2. The temperature dependences of the chemical potential $\tilde{\mu}(\tau)$ for the two-level system.
(*a*) $0 < N \le z_1$:
1— $N = 0.5,\ \tau_{1,2}^{(0.5)} = 5.79,\ \tilde{\mu}_{1,2}^{(0.5)} = 6.01$;
2— $N = 1,\ \tau_{1,2}^{(1)} = 4.21,\ \tilde{\mu}_{1,2}^{(1)} = \tilde{\mu}(0) = g_1^2 = 9.87$;
3— $N = 1.5,\ \tau_{1,2}^{(1.5)} = 3.31,\ \tilde{\mu}_{1,2}^{(1.5)} = 12.08$.
(*b*) $z_1 < N \le z_1 + z_2$:
1— $N = 3,\ \tau_{1,2}^{(3)} = 3.71,\ \tilde{\mu}_{1,2}^{(3)} = 15.43$;
2— $N = 5,\ \tau_{1,2}^{(5)} = 6.88,\ \tilde{\mu}_{1,2}^{(5)} = \tilde{\mu}(0) = g_2^2 = 20.19$;
3— $N = 6,\ \tau_{1,2}^{(6)} = 11.26,\ \tilde{\mu}_{1,2}^{(6)} = 26.76$.

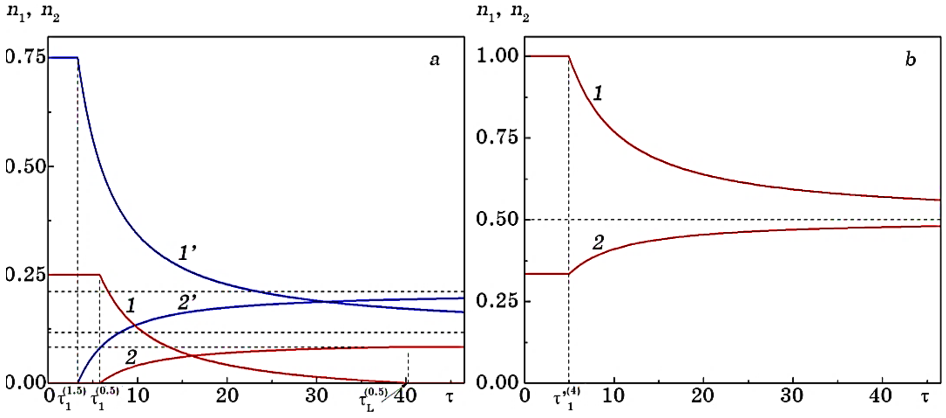

Fig. 3. The temperature dependences of populations for the two-level system.
(*a*) $N = 0.5$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$;
$\tau_{1,2}^{(0.5)} = 5.79,\ n_1 = 0.25,\ n_2 = 0$; $\tau_{01}^{(0.5)} = 40.25,\ n_1 = 0,\ n_2 = 1/12$;
$N = 1.5$: *1'*—$n_1(\tau)$, *2'*—$n_2(\tau)$;
$\tau_{1,2}^{(1.5)} = 3.31,\ n_1 = 0.75,\ n_2 = 0$; $n_{1\infty} = 0.116,\ n_{2\infty} = 0.211$.
(*b*) $N = 0.5$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$;
$\tau_{1,2}^{(4)} = 4.95,\ n_1 = 1,\ n_2 = 1/3$; $n_{1\infty} = n_{2\infty} = 0.5$.

The total energy above temperature $\tau_{1,2}^{(N)}$ in both cases increases monotonically up to some limiting value. The stability condition $\tau - \tau_*(\tau) > 0$ (33) proves to be satisfied at all temperatures.

The calculation of temperature dependences of the heat capacities and other thermodynamic quantities in the two-level system was previously performed by the authors in work [3]. The two-level model is applicable for description of real systems at temperatures below the temperature, at which transitions to the third level begin to occur. At higher temperatures, it is necessary to take into account the contribution to thermodynamics of higher levels as well.

7. THREE-LEVEL SYSTEM

Let us consider a more complex case of the three-level system. For a spherical cavity, the degeneracy factors of the three lower levels are $z_1 = 2,\ z_2 = 6,\ z_3 = 10$. Assume at first that the number of particles is less than the degeneracy factor of the first level, so that the second and third levels at $T = 0$ K are not filled: $N = z_1 n_1 \le z_1,\ n_2 = n_3 = 0$. As the temperature increases, the chemical potential first increases linearly, $\tilde{\mu} = g_1^2 - \tau\theta(N/z_1, z_1)$, but there are no transitions between levels.

Subsequently, at some temperature, there become possible transitions of particles either from the first level to the second one, with

$$\tau_{1,2}^{(N)} = \frac{g_2^2 - g_1^2}{\theta(0, z_2) - \theta(N/z_1, z_1)},\quad \tilde{\mu}_{1,2}^{(N)} = \frac{g_1^2\theta(0, z_2) - g_2^2\theta(N/z_1, z_1)}{\theta(0, z_2) - \theta(N/z_1, z_1)}, \tag{39}$$

or from the first level to the third one, with

$$\tau_{1,3}^{(N)} = \frac{g_3^2 - g_1^2}{\theta(0, z_3) - \theta(N/z_1, z_1)},\quad \tilde{\mu}_{1,3}^{(N)} = \frac{g_1^2\theta(0, z_3) - g_3^2\theta(N/z_1, z_1)}{\theta(0, z_3) - \theta(N/z_1, z_1)}. \tag{40}$$

The sequence of filling of the levels is determined by the relationship between the temperatures $\tau_{1,2}^{(N)}$ and $\tau_{1,3}^{(N)}$. With the available degeneracy factors, it turns out that $\tau_{1,2}^{(N)} < \tau_{1,3}^{(N)}$ for all $N \le z_1$, and so transitions begin to occur from the 1st level to the 2nd one.

When the 1st-level population reaches some minimum value $n_{1\min}$ and the 2nd-level population reaches a maximum value $n_{2\max}$, at $\tau_{12,3}^{(N)}$ and $\tilde{\mu}_{12,3}^{(N)}$, the 3rd level begins to fill up because of transitions from the first two levels. These quantities are found from the set of equations

$$\tilde{\mu}_{12,3}^{(N)} = g_1^2 - \tau_{12,3}^{(N)}\theta\left(n_{1\min}, z_1\right) = g_2^2 - \tau_{12,3}^{(N)}\theta\left(n_{2\max}, z_2\right) = g_3^2 - \tau_{12,3}^{(N)}\theta\left(0, z_3\right), \quad N = z_1 n_{1\min} + z_2 n_{2\max}. \tag{41}$$

At some temperature $\tau_{01}^{(N)}$ for all $N \leq 2$, the population of the first level turns to zero ($n_1 = 0$). This temperature and the corresponding chemical potential are found from the set of equations:

$$\tilde{\mu}_{01}^{(N)} = g_1^2 - \tau_{01}^{(N)}\theta\left(0, z_1\right) = g_2^2 - \tau_{01}^{(N)}\theta\left(n_2, z_2\right) = g_3^2 - \tau_{01}^{(N)}\theta\left(n_3, z_3\right), \quad N = z_2 n_2 + z_3 n_3. \tag{42}$$

With a further increase in temperature, the lower level remains empty and the populations of the other two levels tend to the finite values: $n_{2\infty} = 0.044$, $n_{3\infty} = 0.074$. The temperature dependences of the populations for $N = 1$, as well as of the chemical potential for $N = 1$, $N = 0.5$, $N = 1.5$, are shown in Fig. 4.

At temperatures $\tau < \tau_{12,3}^{(N)}$, the calculations of the temperature de-

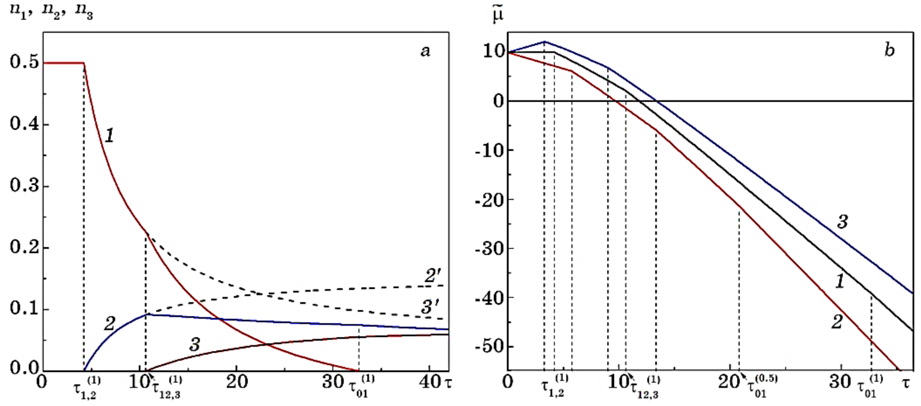

Fig. 4. (*a*) The temperature dependences of the populations for the three-level system with the number of particles $N = 1$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$, *3*—$n_3(\tau)$;
$\tau_{1,2}^{(1)} = 4.21$, $n_1 = 0.5$; $\tau_{12,3}^{(1)} = 10.64$, $n_1 = 0.226$, $n_2 = 0.091$;
$\tau_{01}^{(1)} = 32.73$, $n_1 = 0$, $n_2 = 0.075$, $n_3 = 0.055$; $n_{2\infty} = 0.044$, $n_{3\infty} = 0.074$.
The dashed lines show the calculation of the populations' temperature dependences for the two lower levels in the two-level model.
(*b*) The temperature dependences of the chemical potential $\tilde{\mu}(\tau)$ for the three-level system with the number of particles: *1*—$N = 1$, *2*—$N = 0.5$, *3*—$N = 1.5$;
$\tilde{\mu}_{1,2}^{(1)} = g_1^2 = 9.87$, $\tilde{\mu}_{12,3}^{(1)} = 2.05$, $\tilde{\mu}_{01}^{(1)} = -39.23$;
$\tilde{\mu}_{1,2}^{(0.5)} = 6.01$, $\tilde{\mu}_{12,3}^{(0.5)} = -5.95$, $\tilde{\mu}_{01}^{(0.5)} = -21.38$;
$\tau_{1,2}^{(0.5)} = 5.79$, $\tau_{12,3}^{(0.5)} = 13.37$, $\tau_{01}^{(0.5)} = 20.83$;
$\tilde{\mu}_{1,2}^{(1.5)} = 12.08$, $\tilde{\mu}_{12,3}^{(1.5)} = 6.74$, $\tilde{\mu}_{01}^{(1.5)} = -80.09$;
$\tau_{1,2}^{(1.5)} = 3.31$, $\tau_{12,3}^{(1.5)} = 9.04$, $\tau_{01}^{(1.5)} = 59.97$.

pendences of populations of the two lower levels performed in the two-level and three-level approximations coincide. At higher temperatures, the dependences of populations of the two lower levels in the two-level model shown in Fig. 4, *a* by dashed lines differ from the more accurate calculation in the three-level model. At temperatures $\tau < \tau_{1,2}^{(N)}$, when transitions between levels are yet absent, the chemical potential varies linearly, but the slope of the line depends on the number of particles. For $N < 1$, it is negative, and for $N > 1$, it is positive. With the beginning of transitions of particles from the lower to the upper levels, the chemical potential decreases and becomes negative at high temperatures (Fig. 4, *b*).

Let us now consider the case, when, for the number of particles, $z_1 < N \le z_1 + z_2$, so that, at $T = 0$ K, the first level is completely filled, the second level is partially filled, and the third level is empty: $n_1 = 1,\ n_2 = \left(N - z_1\right)/z_2,\ n_3 = 0$. Depending on the number of particles, there are two possibilities for the system to be excited here. If $N < N_{c1} = 4.21$, then, at $\tau_{1,2}^{(N)}$, the transition of particles from the first level to the second one will begin with the third level being empty (Fig. 5). This point is found from the set of equations

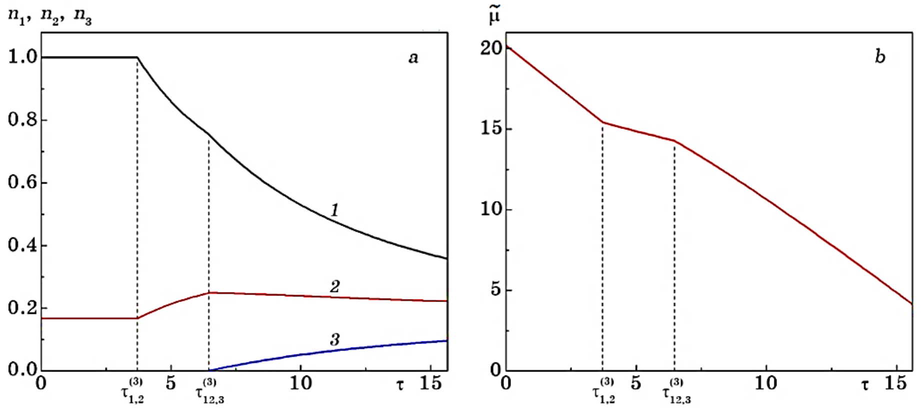

Fig. 5. (*a*) The temperature dependences of populations for the three-level system with $N = 3$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$, *3*—$n_3(\tau)$;
$\tau_{1,2}^{(3)} = 3.71,\ n_1 = 1,\ n_2 = 1/6,\ n_3 = 0$;
$\tau_{12,3}^{(3)} = 6.47,\ n_1 = 0.755,\ n_2 = 0.248,\ n_3 = 0$;
$n_{1\infty} = 0.05,\ n_{2\infty} = 0.167,\ n_{3\infty} = 0.188$.
(*b*) The temperature dependence of the chemical potential $\tilde{\mu}\left(\tau\right)$ for the three-level system with $N = 3$: $\tilde{\mu}\left(0\right) = g_2^2 = 20.19,\ \tilde{\mu}_{1,2}^{(3)} = 15.43,\ \tilde{\mu}_{12,3}^{(3)} = 14.27$.

$$\tilde{\mu}_{1,2}^{(N)} = g_1^2 - \tau_{1,2}^{(N)}\theta(1, z_1) = g_2^2 - \tau_{1,2}^{(N)}\theta(n_2, z_2), \; N = z_1 + z_2 n_2. \tag{43}$$

Subsequently, at the temperature $\tau_{12,3}^{(N)}$ determined from the equations

$$\begin{gathered} \tilde{\mu}_{12,3}^{(N)} = g_1^2 - \tau_{12,3}^{(N)}\theta(n_1, z_1) = g_2^2 - \tau_{12,3}^{(N)}\theta(n_2, z_2) = g_3^2 - \tau_{12,3}^{(N)}\theta(0, z_3), \\ N = z_1 n_1 + z_2 n_2, \end{gathered} \tag{44}$$

because of transitions from the two lower levels, the third level begins to fill up. If $N > N_{c2} = 2.24$, then, in the limit of high temperatures, all three levels have finite populations. And, if $N < N_{c2} = 2.24$, then, at some temperature $\tau_{01}^{(N)}$ determined by the equations

$$\begin{gathered} \tilde{\mu}_{01}^{(N)} = g_1^2 - \tau_{01}^{(N)}\theta(0, z_1) = g_2^2 - \tau_{01}^{(N)}\theta(n_2, z_2) = g_3^2 - \tau_{01}^{(N)}\theta(n_3, z_3), \\ N = z_2 n_2 + z_3 n_3, \end{gathered} \tag{45}$$

the population of the lower level turns to zero.

If $N > N_{c1} = 4.21$, then, at the temperature $\tau_{2,3}^{(N)}$, the transition of particles from the second level to the third one will begin with the first level being completely filled. The point of this transition is determined from the equations

$$\tilde{\mu}_{2,3}^{(N)} = g_2^2 - \tau_{2,3}^{(N)}\theta(n_2, z_2) = g_3^2 - \tau_{2,3}^{(N)}\theta(0, z_3), N = z_1 + z_2 n_2. \tag{46}$$

At the temperature $\tau_{1,23}^{(N)}$ determined from the equations

$$\begin{gathered} \tilde{\mu}_{1,23}^{(N)} = g_1^2 - \tau_{1,23}^{(N)}\theta(1, z_1) = g_2^2 - \tau_{1,23}^{(N)}\theta(n_2, z_2) = g_3^2 - \tau_{1,23}^{(N)}\theta(n_3, z_3), \\ N = z_1 + z_2 n_2 + z_3 n_3, \end{gathered} \tag{47}$$

the transition of particles from the lower level to the second and third levels begins. The temperature dependences of populations and chemical potential for this case are shown in Fig. 6.

The critical values N_{c1} and N_{c2} are found from the equations $\tau_{1,2}^{(N_{c1})} = \tau_{2,3}^{(N_{c1})}$ and $1/\tau_{01}^{(N_{c2})} = 0$.

Let us finally consider the possibility, when, for the number of particles, $z_1 + z_2 < N \le z_1 + z_2 + z_3$, so that, at $T = 0$ K, the first and second levels are completely filled, and the third level is partially filled: $n_1 = 1, n_2 = 1, n_3 = (N - z_1 - z_2)/z_3$. In this case, for all N at the available degeneracy factors, there begin transitions from the second level to the third one at the temperature $\tau_{2,3}^{(N)}$ determined from the equations

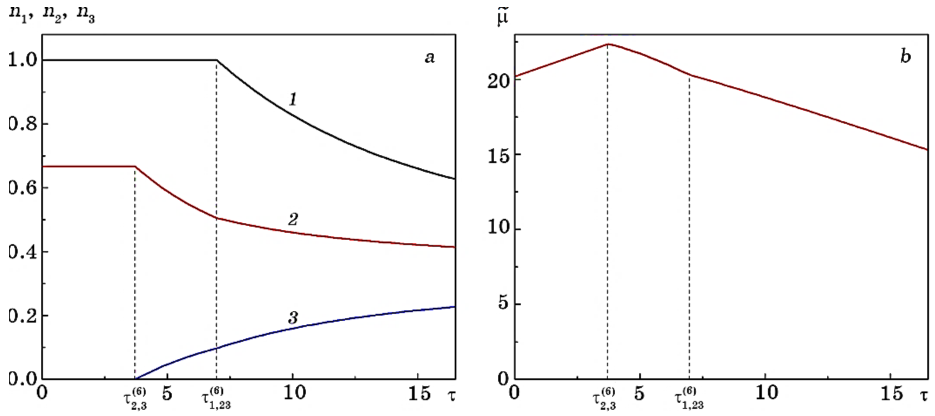

Fig. 6. (*a*) The temperature dependences of populations for the three-level system with $N = 6$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$, *3*—$n_3(\tau)$;
$\tau_{2,3}^{(6)} = 3.71,\ n_1 = 1,\ n_2 = 2/3,\ n_3 = 0$;
$\tau_{1,23}^{(6)} = 6.96,\ n_1 = 1,\ n_2 = 0.505,\ n_3 = 0.097$;
$n_{1\infty} = 0.280,\ n_{2\infty} = 0.334,\ n_{3\infty} = 0.344$.
(*b*) The temperature dependence of the chemical potential $\tilde\mu(\tau)$ for the three-level system with $N = 6$: $\tilde\mu(0) = g_2^2 = 20.19,\ \tilde\mu_{2,3}^{(6)} = 22.35,\ \tilde\mu_{1,23}^{(6)} = 20.32$.

$$\tilde\mu_{2,3}^{(N)} = g_2^2 - \tau_{2,3}^{(N)}\theta(1, z_2) = g_3^2 - \tau_{2,3}^{(N)}\theta\big((N - z_1 - z_2)/z_3, z_3\big). \qquad (48)$$

At the higher temperature $\tau_{1,23}^{(N)} > \tau_{2,3}^{(N)}$ determined from the set of equations

$$\begin{gathered}\tilde\mu_{1,23}^{(N)} = g_1^2 - \tau_{1,23}^{(N)}\theta(1, z_1) = g_2^2 - \tau_{1,23}^{(N)}\theta(n_2, z_2) = g_3^2 - \tau_{1,23}^{(N)}\theta(n_3, z_3),\\ N = z_1 + z_2 + z_3 n_3,\end{gathered} \qquad (49)$$

there also begin transitions of particles from the first level to the second and third levels. The temperature dependences of populations and chemical potential for this case with $N = 9$ are shown in Fig. 7.

In the case, when the number of particles exceeds the critical value, $N > N_{c3} = 15.76$, the transitions from the lower level become impossible, so that, at all temperatures, $n_1 = 1$, and there are transitions only from the second level to the third one. Note as well that, at a small number of particles, $N < N_{c4} = 0.335$, at the temperature

$$\tau_{012}^{(N)} = \frac{g_3^2 - g_2^2}{\theta(N/z_3, z_3) - \theta(0, z_2)}, \qquad (50)$$

the two lower levels become empty and only the upper level is populated. The critical values N_{c3} and N_{c4} are found from the equations

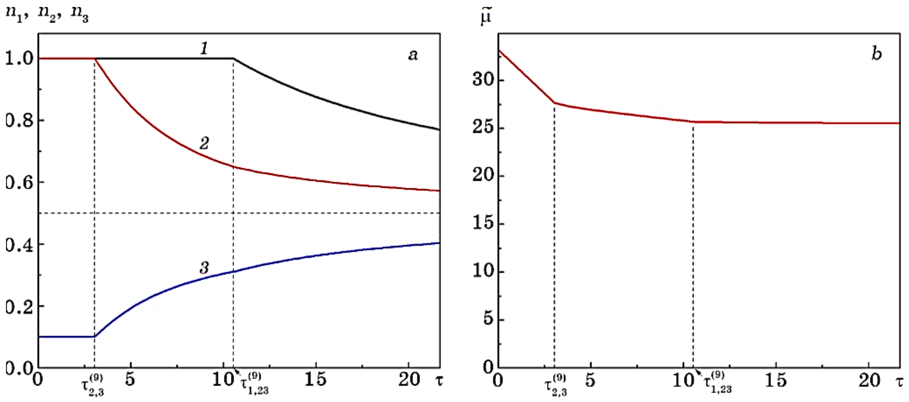

Fig. 7. (*a*) The temperature dependences of populations for the three-level system with $N = 9$: *1*—$n_1(\tau)$, *2*—$n_2(\tau)$, *3*—$n_3(\tau)$;
$\tau^{(9)}_{2,3} = 3.04,\ n_1 = 1,\ n_2 = 1,\ n_3 = 0.1$;
$\tau^{(9)}_{1,23} = 10.53,\ n_1 = 1,\ n_2 = 0.649,\ n_3 = 0.310$;
$n_{1\infty} = n_{2\infty} = n_{3\infty} = 0.5$.
(*b*) The temperature dependence of the chemical potential $\tilde{\mu}(\tau)$ for the three-level system with $N = 9$: $\tilde{\mu}(0) = g_3^2 = 33.22,\ \tilde{\mu}^{(9)}_{2,3} = 27.65,\ \tilde{\mu}^{(9)}_{1,23} = 25.67$.

$1/\tau^{(N_{c3})}_{1,23} = 0$ and $1/\tau^{(N_{c4})}_{012} = 0$. As the temperature increases, more and more levels become populated and have to be taken into account. In this case, the calculations become more complicated, but do not principally differ from the considered cases.

Knowing temperature dependences of the level populations, one can calculate thermodynamic quantities by means of the general formulas (22)–(26). Figure 8 shows the result of calculation of the heat capacities in the three-level model with $N = 1$. As we can see, at temperatures corresponding to the beginning of a new level filling, the heat capacity experiences a jump and then gradually decreases. The magnitude of the jumps decreases with increasing temperature. There is also a jump in the heat capacity at the temperature $\tau^{(1)}_{01}$, when the population of the lower level turns to zero.

Let us estimate the temperature and the sphere radius, at which the heat-capacity jumps can be observed. The temperature in dimensional units T is related to the dimensionless temperature τ, according to (28), by the relation

$$T = \tau\frac{\hbar^2}{2ma^2}, \tag{51}$$

so that $T = 0.43\cdot 10^{-11}\,\tau/a^2$ $[\mathrm{K}\cdot\mathrm{cm}^2]$. The above-calculated tempera-

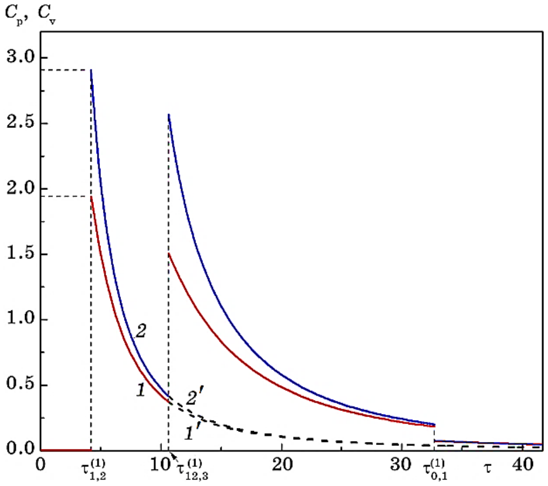

Fig. 8. The temperature dependences of the heat capacities for $N = 1$: *1*—$C_V(\tau)$, *2*—$C_p(\tau)$. The dashed line at $\tau > \tau^{(1)}_{12,3}$ shows the calculation in the two-level model. At $\tau \le \tau^{(1)}_{12,3}$, the dependences $C_V(\tau)$, $C_p(\tau)$ for the two- and three-level models coincide. The magnitudes of jumps are as follow:
$\Delta C_V\left(\tau^{(1)}_{1,2}\right) = 1.94$, $\Delta C_p\left(\tau^{(1)}_{1,2}\right) = 2.91$; $\Delta C_V\left(\tau^{(1)}_{12,3}\right) = 1.13$, $\Delta C_p\left(\tau^{(1)}_{12,3}\right) = 2.15$;
$\Delta C_V\left(\tau^{(1)}_{01}\right) = -0.11$, $\Delta C_p\left(\tau^{(1)}_{01}\right) = -0.12$.

tures, at which the transition of particles from the ground level to the next level begins for different N, have close values from several to ten units: $\tau^{(0.5)}_{1,2} = 5.79$, $\tau^{(1)}_{1,2} = 4.21$, $\tau^{(1.5)}_{1,2} = 3.31$, $\tau^{(3)}_{1,2} = 3.71$, $\tau^{(4)}_{1,2} = 4.95$, $\tau^{(5)}_{1,2} = 6.88$, $\tau^{(6)}_{1,2} = 11.26$. Let us take for estimations $\tau \approx 10$. Then, in order to observe a jump in the heat capacity at $T = 1$ K, the radius should be $a \approx 0.7 \cdot 10^{-5}$ cm. At present, temperatures of the order of 10^{-3} K are achieved. In this case, $a \approx 2 \cdot 10^{-4}$ cm. Thus, the observation of jumps in heat capacities is possible for particles of macroscopic sizes.

The possibility of observing size effects for the heat capacity owing to discreteness of levels in colloidal metal solutions was discussed yet in the early work of Fröhlich [4]. He noted that electron levels broaden because of the interaction of electrons with phonons. Therefore, for the discrete level structure to be clearly expressed, the size of the metal particles must be smaller than the free path length of electrons. Currently, colloidal solutions of metal nanoparticles with sizes in the range from 1 nm to 15 μm are created and experiments can be carried out at temperatures close to millikelvin. Thus, there is a hope that the prediction of the heat capacity jumps in small particles can be verified experimentally.

With increasing the radius a, the distance between levels decreases, so that, in the limit $a \to \infty$, the transition to the continuum limit becomes possible [5]. In this limit, the formulae obtained in this work turn into the known formulae for the thermodynamic quantities of the Fermi gas [9].

8. CONCLUSIONS

In this paper, the thermodynamics of fermions in a sphere of an arbitrary radius with an arbitrary and even small number of particles is studied. The work is a development of work [5], where the thermodynamics of Fermi particles in a cubic cavity is considered. The main attention is paid to the study of size effects due to the discrete structure of energy levels. Since the structure of levels and the degeneracy multiplicity of levels in the sphere are different from their structure and multiplicity in the cubic cavity, the results are quantitatively different from the results of work [5]. On the examples of the two- and three-level models, the dependences of the level populations and the chemical potential on temperature are considered. It is shown that at temperatures corresponding to the beginning of a new level filling, the heat capacity experiences jumps. The magnitude of the jumps decreases with increasing temperature.

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