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**THERMODYNAMICS OF METALLIC HELIUM**

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*The internal and free energies of liquid metallic helium are calculated for wide ranges of density and temperature, and the corresponding equation of state is obtained in the framework of perturbation theory. The electron-ion interaction potential is selected as a small parameter, and the calculations are carried out to the third order of smallness inclusive. Conduction electrons are considered in the random phase approximation with regard for the exchange interaction and correlations in the local field approximation. The hard-sphere model is used for the nuclear subsystem, the sphere diameter being the only parameter of the theory. The sphere diameter and the system density, at which helium transforms from the single- into double-ionized state are evaluated by analyzing the effective pair interaction between helium nuclei also in the third order of perturbation theory. The case of double-ionized helium atoms is considered. The third-order correction turns out substantial in all examined cases. The values obtained for thermodynamic parameters such as the density, temperature, and pressure fall within the ranges typical of the central regions of giant planets, which allows us to suppose the existence of metallic helium in the solar system.*

*Keywords:* liquid metallic helium, thermodynamic parameters, giant planets.

**1. Introduction**

Progress in the experiments dealing with the shock compression of substances allowed one to metallize those of them that are gases in the normal state [1, 2]. In particular, hydrogen and deuterium in the metallic state were obtained and studied in details in 1996–1999 [3–5], oxygen in 2001 [6], and nitrogen in 2003 [7]. However, nobody has succeeded in obtaining helium in the metallic state experimentally till now. Nevertheless, the theoretical study of the process of probable helium metallization runs more productively, although the total number of works devoted to this subject is rather small.

According to one of the first relevant works [8], a pressure of 100 Mbar is required for helium metallization at low temperatures, which makes its ex-

istence in the metallic state unreal even in the internal regions of giant planets in the solar system. Within the quantum-mechanical method of molecular dynamics, the authors of work [9] obtained rather an unexpected result that the metal–insulator transition can occur in liquid helium already at a density of 1 g/cm<sup>3</sup>. In work [10], the quantum-mechanical Monte-Carlo method was used to obtain the equation of state for solid helium. The corresponding results consist in that the metal–insulator transition has to take place at a density of 21.3 g/cm<sup>3</sup> and a pressure of 25.7 Mbar. In work [11], the equation of state and the coefficient of electroconductivity for helium were calculated in the framework of the molecular dynamics method and with the use of the density functional method for the electron subsystem. The cited authors claimed that the temperature is a crucial factor for the helium electron structure. In particular, the

energy gap disappears at a density of  $13 \text{ g/cm}^3$  at the zero temperature and at  $6.6 \text{ g/cm}^3$  at a temperature of  $20,000 \text{ K}$ . The latter values are quite reachable in Jupiter's internal region.

As follows from the aforesaid, the calculations of various properties of metallic helium have been carried out with the use of mathematical simulation methods, and the results obtained are not in complete agreement with one another. This work is one of the first ones devoted to the application of analytical methods to studying the properties of metallic helium, and its main aim is to study the corresponding equation of state. The theory contains a single fitting parameter, namely the diameter of solid spheres. To determine it, the effective ion-ion pair interaction will be analyzed by analogy with the cases of metallic hydrogen and other simple metals [12–16].

## 2. Internal Energy

While studying metallic helium, let us take advantage of the Hamiltonian [13, 17] used for the analysis of simple liquid metals, which takes the electron-electron interaction into account exactly. The internal energy of the metal can be obtained by averaging the Hamiltonian over the Gibbs canonical ensemble

$$E = E_i + E_{ie}. \quad (1)$$

Here,

$$E_i = N_i \frac{3}{2} k_B T + N_i \frac{1}{2V} \sum_{\mathbf{q}}' V_{ii}(q) [S_i(q) - 1] \quad (2)$$

is the contribution of the nuclear subsystem. The first term on the right-hand side of equality (2) is the kinetic energy of nuclei, the second one corresponds to the Coulomb energy of their interaction,  $V$  is the system volume,  $N_i$  the number of nuclei,  $V_{ii}(q) = 4\pi z^2 e^2 / q^2$  is the Fourier transform of the energy of Coulomb interaction between helium nuclei,  $z = 2$ ,  $T$  is the absolute temperature, the prime means the absence of the term with  $\mathbf{q} = 0$ , and  $S_i(q)$  is the static structure factor of the nuclear subsystem. In what follows, the structure factor calculated in the hard-sphere model will be used for  $S_i(q)$ . For a given concentration, it depends parametrically only on the solid sphere diameter. At the temperatures considered below, the nuclear subsystem can be regarded as classical.

The energy of the electron subsystem and the energy of interaction between the electron and nuclear subsystems are convenient to be analyzed together. Their sum, being the energy of the ground state of the electron gas in the field of nuclei, can be expanded in a series in the electron-nucleus interaction parameter [13]

$$E_{ie} = \sum_{n=0}^{\infty} E_n. \quad (3)$$

Every term in this series, in turn, should be expanded in a series in the electron-electron interaction parameter. The term of the zeroth order in the electron-nucleus interaction, when using the Nozières–Pines interpolation formula [18, 19], looks like

$$E_0 = N_e \left( \frac{1.105}{r_s} - \frac{0.458}{r_s} - 0.058 + 0.016 \ln r_s \right). \quad (4)$$

Here, the Brueckner non-ideality parameter  $r_s$  was introduced, which equals the radius of a sphere, the volume of which is equal to a volume per one electron, and  $N_e$  is the number of electrons in the metal.

Owing to the electroneutrality of the system, the first-order correction in the electron-electron interaction to the ground-state energy of the electron gas in metallic helium is absent. The terms of the second and higher orders look like

$$E_n = \frac{N_i}{V^n} \sum_{\mathbf{q}_1, \dots, \mathbf{q}_n} \Gamma^{(n)}(\mathbf{q}_1, \dots, \mathbf{q}_n) V_{ie}(q_1) \dots V_{ie}(q_n) \times \\ \times S_i(\mathbf{q}_1, \dots, \mathbf{q}_n) \Delta(\mathbf{q}_1 + \dots + \mathbf{q}_n). \quad (5)$$

Here,  $V_{ie}(q) = 4\pi z e^2 / q^2$  is the Fourier transform of the Coulomb electron-nucleus interaction energy,  $S_i(\mathbf{q}_1, \dots, \mathbf{q}_n)$  is the  $n$ -particle structure factor of the nuclear subsystem;  $\Delta(\mathbf{q}_1 + \dots + \mathbf{q}_n)$  the Kronecker symbol, and  $\Gamma^{(n)}(\mathbf{q}_1, \dots, \mathbf{q}_n)$  the electron  $n$ -pole factor [13]. The last expression is formally exact and, hence, is unsuitable for specific calculations.

There are a number of techniques to approximately calculate the electron three-pole factor [20–23] and many-particle structure factors for the nuclear subsystem [24]. For the electron double-pole factor, the general result obtained by various authors reads

$$\Gamma^{(2)}(\mathbf{q}, -\mathbf{q}) = -\frac{1}{2} \frac{\pi(q)}{\varepsilon(q)}. \quad (6)$$

Here,  $\pi(q)$  is the polarization function, and  $\varepsilon(q)$  is the dielectric permittivity of the electron gas. In the random phase approximation with regard for the exchange interaction and electron–electron correlations in the local-field approximation, we have

$$\varepsilon(q) = 1 + [V_{ee}(q) + U(q)]\pi_0(q), \quad (7)$$

where  $V_{ee}(q) = 4\pi e^2/q^2$  is the Fourier transform of the Coulomb electron–electron interaction energy,  $U(q) = -2\pi e^2/(q^2 + \lambda k_F^2)$  is the potential energy of exchange interaction and correlations in the electron gas,  $\lambda \approx 2$  [25], and  $\pi_0(q)$  is the polarization function of the ideal electron gas.

The results obtained by different authors for the electron three-pole factor are substantially different. The result of our independent calculations [17] carried out in the framework of the ideal electron gas model coincides with that obtained in work [22]:

$$\Gamma^{(3)}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3) = \frac{\Lambda_0^{(3)}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)}{\varepsilon(\mathbf{q}_1)\varepsilon(\mathbf{q}_2)\varepsilon(\mathbf{q}_3)}. \quad (8)$$

Here,  $\Lambda_0^{(3)}(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3)$  is the electron three-pole factor for a degenerate ideal electron gas. After changing from summation to integration in a spherical coordinate system, the terms of the second and third orders in the electron–proton interaction are as follows:

$$E_2 = -N_i \frac{1}{4\pi^2} \int_0^\infty \frac{\pi(q)}{\varepsilon(q)} V_{ie}^2(q) S_i(q) q^2 dq, \quad (9)$$

$$E_3 = N_i \frac{1}{4\pi^4} \int_0^\infty dq_1 q_1^2 \int_0^\infty dq_2 q_2^2 F(q_1, q_2), \quad (10)$$

$$F(q_1, q_2) = \frac{2n + 1}{2} \int_0^\pi \frac{\Lambda_0^{(3)}(\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_1 - \mathbf{q}_2)}{\varepsilon(q_1)\varepsilon(q_2)\varepsilon(|\mathbf{q}_1 - \mathbf{q}_2|)} \times \\ \times V_{ie}(q_1)V_{ie}(q_2)V_{ie}(|\mathbf{q}_1 + \mathbf{q}_2|)S_i(\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_1 - \mathbf{q}_2) \times \\ \times \sin(\theta_{12})d\theta_{12}. \quad (11)$$

For the three-particle structure factor, we use the approximation [24, 26, 27]

$$S_i(\mathbf{q}_1, \mathbf{q}_2, \mathbf{q}_3) = S_i(\mathbf{q}_1)S_i(\mathbf{q}_2)S_i(\mathbf{q}_3). \quad (12)$$

The Helmholtz free energy is defined as

$$F = E - TS, \quad (13)$$

where  $S$  is the entropy of the system. It is expressed as a sum of electron and nuclear components. In the case of degenerate electron gas, the electron component can be neglected in comparison with the nuclear one. The latter contribution, in the hard-sphere approximation [19, 28], looks like

$$S = S_{0i} + S_{ii}(\eta), \quad (14)$$

where

$$S_{0i} = N_i k_B \ln \left[ \frac{e}{n_i} \left( \frac{eMk_B T}{2\pi\hbar^2} \right)^{3/2} \right] \quad (15)$$

is the entropy of the ideal nuclear gas,  $M$  the nucleus mass,  $n_i$  the concentration of nuclei,

$$S_{ii}(\eta) = N_i k_B \frac{3\eta^2 - 4\eta}{(1 - \eta)^2} \quad (16)$$

is the contribution associated with the interaction between nuclei, and  $\eta$  is the parameter of nuclear packing.

### 3. Effective Ion-Ion Pair Interaction

At first glance, the proposed theory contains a single undetermined parameter, the diameter of hard spheres. Knowing its dependence on the concentration and the temperature, the corresponding dependences of thermodynamic potentials can also be found. In this work, in order to determine the required dependence for the hard-sphere diameter, we use the approximation of effective pair interaction between nuclei. The expression for this interaction includes the direct Coulomb interaction between nuclei and their interaction mediated through conduction electrons. The former is repulsive, the latter attractive. The competition between those two components may result in a formation of a potential well with a characteristic minimum. From the dependence of this interaction on the distance between nuclei, it is possible to determine the diameter of hard spheres for any temperature and concentration [29].

The diameter of hard spheres  $\sigma$  is a minimum distance, at which the nuclei can approach one another at a given temperature. It can be found from the condition of equality of the kinetic and potential energies of nuclei at their mutual approach,

$$V_{ef}(\sigma) = 3k_B T/2. \quad (17)$$

Analogously to the case of metallic hydrogen [12, 30, 31], the effective ion-ion pair interaction in liquid metallic helium is considered in the framework of the theory of perturbation in the electron-nucleus interaction. The quantity concerned can be expressed as follows [13]:

$$U_{\text{ef}}(R) = \sum_{n=0}^{\infty} U_2^{(n)}(R), \quad (18)$$

where  $R$  is the distance between nuclei. If metallic helium is formed by double-ionized helium atoms,  $U_2^{(0)}(R) = V_{ii}(R)$  is the energy of Coulomb interaction between nuclei. The term of the first order in the electron-nucleus interaction potential  $U_2^{(1)}(R) = 0$ , the term of the second order is

$$U_2^{(2)}(R) = -\frac{e^2}{2\pi^2 R} \int_0^{\infty} V_{ei}^2(q) \frac{\pi_0(q)}{\varepsilon(q)} \sin(qR) q dq, \quad (19)$$

and the term of the third order is

$$U_2^{(3)}(R) = \frac{3e^2}{4\pi^4 R} \int_0^{\infty} dq_1 q_1 \sin(q_1 R) \int_0^{\infty} dq_2 q_2^2 \int_{|q_1 - q_2|}^{q_1 + q_2} dq_3 \times \\ \times \frac{V_{ie}(q_1) V_{ie}(q_2) V_{ie}(q_3)}{\varepsilon(q_1) \varepsilon(q_2) \varepsilon(q_3)} \Lambda_0^{(3)}(q_1, q_2, q_3). \quad (20)$$

In Fig. 1, the plot of the dependence of the effective pair interaction between nuclei on the distance between them is depicted. The potential is reckoned in kelvins, and the distance in atomic units. The potential has a profile typical of that for simple metals. The depth of the first potential well minimum amounts to approximately 3000 K, which is enough for liquid metallic helium to be stable at lower temperatures and the given density. The position of the first minimum corresponds to the equilibrium relative arrangement of neighbor helium nuclei in the metal phase. The term of the third order has much more importance to the formation of this minimum than the second-order term does. Such a situation is inherent to metallic hydrogen as well [15]. Note that, nevertheless, the contribution of the third-order term to the internal energy of the metal amounts only to a few percent, being much smaller than the contribution of the second-order term.

The depth of the first minimum decreases at higher densities and increases at lower ones. In addition, another potential well starts to emerge at much shorter

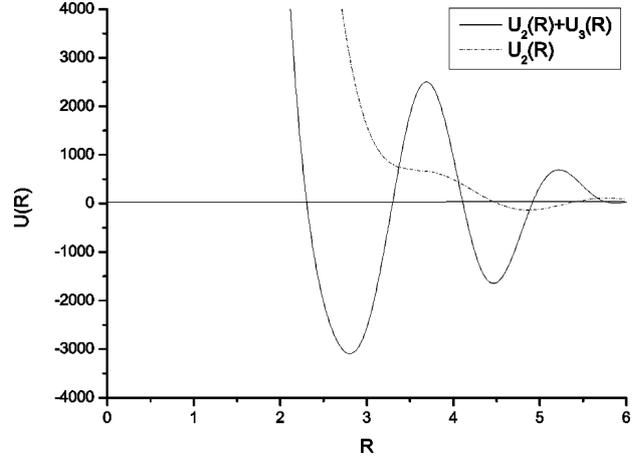
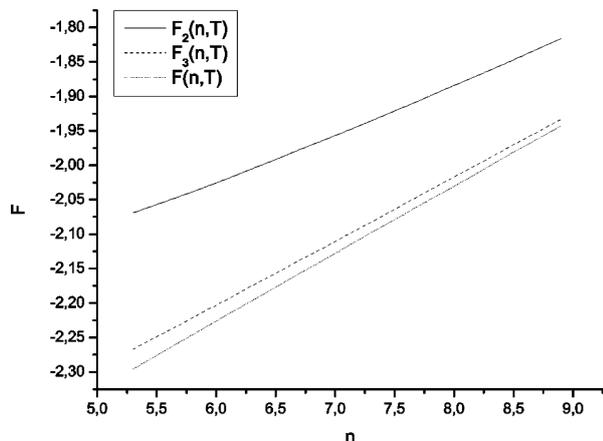


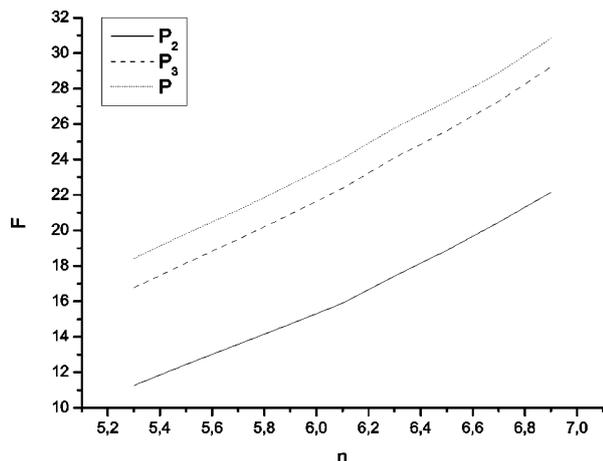
Fig. 1. Potential of effective pair interaction between helium nuclei at a density of 6.7 g/cm<sup>3</sup>

distances between the nuclei. Such a behavior of the potential of proton-proton interaction is also observed in metallic hydrogen. It is usually interpreted as the beginning of the formation of the molecular hydrogen phase [30, 31]. In the case of helium, we, probably, also obtain a reconstruction of the electron subsystem in the metal, which corresponds to the emergence of single-ionized helium atoms. In our opinion, the characteristic density that separates those two metallic phases of liquid helium is a density of 5.3 g/cm<sup>3</sup>, at which the depths of two indicated potential wells become equal. The situation with metallic helium is unique. We can calculate only the free energy of double-ionized helium. That is why our analysis is based on the effective pair interaction rather than, for instance, the free energy in a vicinity of the transition point. The formulas for the pair interaction do not contain any free parameter and do not demand any grave model assumptions. The analysis of any other metallic helium parameter would require the both.

The results of our calculations testify that helium can be in the metallic state in the central regions of all giant planets in the solar system. Note that the results obtained, while finding the hard sphere diameter, should be regarded only as useful estimations. The issue concerning the role of terms in the perturbation theory series higher than the third-order one for the calculations of the effective pair nucleus-nucleus interaction remains open. In particular, for a temperature of 10,000 K and a helium density of 5.3 g/cm<sup>3</sup>, we obtain 2.101 a.u. for the diameter of

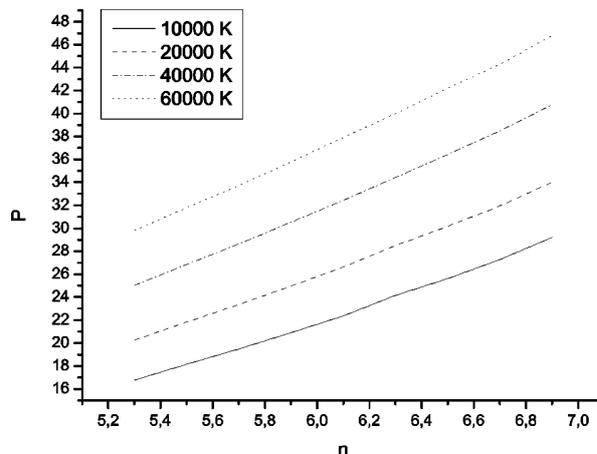


**Fig. 2.** Dependences of the Helmholtz free energy on the density at  $T = 10000$  K found in the second,  $F_2(n, T)$ , and third,  $F_3(n, T)$ , orders of perturbation theory, and an estimation of the total sum for  $F(n, T)$

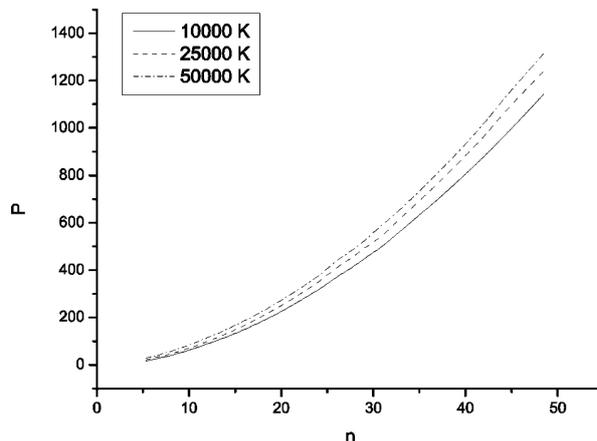


**Fig. 3.** Dependences of the pressure on the density at  $T = 10000$  K found in the second,  $P_2(n, T)$ , and third,  $P_3(n, T)$ , orders of perturbation theory, and an estimation of the total sum for  $P(n, T)$

hard spheres in the second order of perturbation theory. If the third-order term is taken into consideration, this value falls down to 1.778 a.u. The terms of the fourth and higher orders cannot be taken into account accurately. We may only suppose that including the terms of higher orders into consideration would result in a further reduction of the hard-sphere diameter. If we additionally suppose that the rate of those variations corresponds to the geometric progression, the value of hard-sphere diameter obtained



**Fig. 4.** Dependences of the pressure on the density calculated in the third order of perturbation theory for various temperatures



**Fig. 5.** The same as in Fig. 4, but for very high densities

in the second order of perturbation theory should be multiplied by  $\alpha = 0.5$ . In this case, it turns out close to the diameter of the first Bohr orbit in a single-ionized helium atom.

#### 4. Discussion of Results

First of all, let us elucidate how eligible is the application of perturbation theory in the electron-nucleus interaction to the determination of the free energy. In Fig. 2, the dependence of the free energy on the density is exhibited. The free energy  $F(n, T)$  was estimated, by assuming that the series of perturbation theory for the structure-dependent part of the internal energy is a geometric progression with the com-

mon ratio equal to  $E_3(n, T)/E_2(n, T)$ . The energy values are reckoned in atomic units, and the density in  $\text{g}/\text{cm}^3$  units. One can see that the obtained estimation is close to the values of energy calculated to within the third-order terms of perturbation theory.

A similar behavior is inherent to the dependence of the pressure on the density, which is illustrated in Fig. 3. The pressure is reckoned in Mbar units and the density in  $\text{g}/\text{cm}^3$  ones. As the density grows, the convergence rate for the free energy and pressure series increases. At a density of  $5.3 \text{ g}/\text{cm}^3$ , the pressure amounts to about 18 Mbar. Provided that helium is in the single-ionized state below this density, the indicated pressure value may probably be the upper pressure limit for helium in this state. The values of pressure and density shown in Fig. 3 fall within the corresponding ranges that are characteristic of the inner regions of such planets as the Jupiter [32]. This circumstance points at a possibility for metallic helium to exist in the central regions of not only the Jupiter, but also other exoplanets belonging to its group.

In Fig. 4, the pressure is plotted as a function of the density at various temperatures. This dependence grows monotonously, being almost linear in the considered intervals of densities and temperatures. The pressure is reckoned in Mbar units, and the density in  $\text{g}/\text{cm}^3$  ones.

Figure 5 demonstrates the dependence of the pressure on the density for so high values of the latter that the pressure no more depends on the structure of the nuclear subsystem. At the end of the considered interval, the results of summing up the series of perturbation theory for the pressure including the terms of the second or third order coincide to within three significant digits,  $P_2(50, 10000) = P_3(50, 10000)$ . The relative magnitude of temperature corrections also decreases. The behavior of the pressure becomes more and more universal, which is mainly governed by properties of the degenerate electron gas.

The reliability of the obtained results is also confirmed by the fact that the electrical resistivity of metallic helium calculated similarly to that of metallic hydrogen [15] falls within the interval of values typical of simple bivalent liquid metals. Its temperature dependence is also similar to that for metallic hydrogen.

The results of our calculations provide quite reasonable values, which may serve as a reference point, while studying the thermodynamic properties

of single-ionized helium. In this case, the developing theory will inevitably contain more fitting parameters, so that experimental information associated with the metallic state of helium will be required for their determination.

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#### ТЕРМОДИНАМІКА МЕТАЛІЧНОГО ГЕЛІЮ

#### Резюме

В роботі розраховано внутрішню і вільну енергію, та рівняння стану в рідкому металічному гелії в широкому діапазоні густин і температур. Для всіх зазначених характе-

ристик металу використана теорія збурень за потенціалом електрон-іонної взаємодії із врахуванням членів до третього порядку включно. Для електронів провідності використано наближення випадкових фаз при врахуванні обмінної взаємодії і кореляцій в наближенні локального поля. Для ядерної підсистеми використана модель твердих сфер. Діаметр цих сфер є єдиним параметром теорії. Оцінку діаметра і густини системи, за яких відбувається перехід гелію з одноразово у дворазово іонізований стан, виконано на основі аналізу парної ефективної взаємодії між ядрами гелію. Для останньої також враховується член третього порядку теорії збурень. В роботі розглянуто випадок дворазово іонізованих атомів гелію. Роль поправки третього порядку у всіх випадках виявилась суттєвою. Значення термодинамічних параметрів: густини, температури і тиску укладаються в діапазон значень, характерних для центральних областей планет-гігантів. Це дозволяє припустити існування гелію в металічному стані в межах сонячної системи.