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ON STABILITY OF NANOSCALE ELECTRON-POSITRON DROPLETS

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О СТАБИЛЬНОСТИ НАНОРАЗМЕРНЫХ ЭЛЕКТРОН-ПОЗИТРОННЫХ КАПЕЛЬ

This paper presents the results of the total energy calculation for electroneutral electron-positron clusters with closed shells. The calculations were performed within the Random Phase Approximation with Exchange to take into account many-particle correlations that allowed us to reduce the energy per one electron-positron pair below the energy per one pair of dipositronium molecule. The most stable electron-positron clusters are found for the cluster sizes in the interval from 20 to 40 pairs.

CLUSTER, ELECTRON, POSITRON, NANOSCALE DROPLET, RPAE.

В статье представлены результаты вычислений полных энергий электронейтральных электрон-позитронных кластеров с заполненными оболочками. Для учета многочастичных корреляций вычисления проводились в рамках приближения случайных фаз с обменом, которое позволило получить энергию на одну электрон-позитронную пару меньше той же энергии для молекулы дипозитрония. Обнаружено, что наиболее стабильными являются кластеры, содержащие число пар в интервале от 20 до 40.

КЛАСТЕР, ЭЛЕКТРОН, ПОЗИТРОН, НАНОРАЗМЕРНАЯ КАПЛЯ, ПСФО.

I. Introduction

Investigations of a bound state of matter and antimatter, in particular of electron-positron systems, have a long story. Before the experimental observation of the atom-like bound state of an electron and positron in 1951 [1], the probability of formation of the dipositronium molecule – the bound state of two positroniums – was predicted theoretically in 1946 [2]. Then, in 1947, the ground state energy of dipositronium molecule was calculated by solving the four-body Shrodinger equation for two electrons and two positrons [3]. Later on, a set of computations of coupling energy of diposi-

tronium was performed and the lifetime of that system was estimated as well [4 – 6]. Finally, the dipositronium molecule was found experimentally in 2005 [7] that proved the existence of bound state of two electron-positron pairs.

The problem of possible formation of compact bound systems of more than two electron-positron pairs is still open. Recently, it was shown that formation of an electron-positron liquid is possible in particular interval of particle densities and temperatures due to intense Coulomb interaction that correlates the motion of electrons and positrons [8]. So far, one could have expected a formation of electron-

positron droplets similar to the electron-hole droplets in semiconductors [9]. The number of particles in the droplets theoretically can vary from two electron-positron pairs to infinity. In the similar way, as it happens in positronium atom, the motion of electrons and positrons in a finite-size droplets must be quantized. Electrons and positrons move in the self-consistent field formed by both subsystems with the whole system being electrically neutral. It was proposed that the droplets formed by mutual interaction between electron and positron subsystems should have a lot of similarities with alkali metal clusters, where their valence electrons are completely delocalized through the whole volume. Based on this proposal, a new theoretical model for electron-positron droplets was developed [10 – 12]. In particular, in the papers [10 – 12] the electron-positron droplets containing up to 100 particle pairs have been studied. Calculations of internal structure of these systems were performed both within Hartree – Fock (HF) approximation and the Local Density Approximation (LDA). It was found that similarly to metallic clusters [13], the minimal energies per one particle correspond to the systems with «magic» numbers of electron-positron pairs that form closed shells.

It was shown in [10 – 12] that the optimal density distribution of both subsystems corresponding to an absolute minimum of the total energy of electron-positron droplet must satisfy the local electroneutrality condition that provides the local equality of positive and negative charge densities through the whole droplet volume. The similar situation takes place with alkali metal clusters within the optimized jellium model [14].

The previous calculations [10 – 12] show that the energies per one pair resulting from the latter calculations are significantly higher than the energy per pair in dipositronium molecule [4 – 6]. However, one should note that HF and LDA approximations used in these calculations do not take into account the dynamical part of interparticle correlations, in particular the polarization interaction. Moreover, the energies of electron-positron clusters obtained within the density functional theory framework for a part of many-particle correlations [10, 11] are visibly lower than corresponding Hartree-Fock energies [12].

The goal of present work is to study the role of correlation effects in possible formation of stable electron-positron clusters. The correlation corrections to the Hartree-Fock coupling energies of several most stable clusters with «magic» numbers of pairs were calculated using different approximations. The results of preliminary calculations were already presented in [15]. The present paper is devoted to more detailed description of obtained results.

The atomic system of units is used throughout the paper: $\hbar = |e| = m_e = 1$.

II. Theoretical Approach

Let's consider an electrically neutral system of fermions interacting via Coulomb forces. For example, it can be an electron-positron droplet which consists of the same number N of positive and negative charged fermions with equal masses $m_e = m_p = m = 1$. The ground state wave function of the system described by total Hamiltonian \hat{H}_0 with pair interaction potential

$$V(\mathbf{r}, \mathbf{r}') = \frac{1}{|\mathbf{r} - \mathbf{r}'|},$$

$$\hat{H}_0 = -\sum_{i=1}^N \frac{\Delta_i}{2} - \sum_{a=1}^N \frac{\Delta_a}{2} + \sum_{i \neq j} V(\mathbf{r}_i^{(e)} - \mathbf{r}_j^{(e)}) + \sum_{a \neq b} V(\mathbf{r}_a^{(p)} - \mathbf{r}_b^{(p)}) + \sum_{i,a} V(\mathbf{r}_i^{(e)} - \mathbf{r}_a^{(p)}), \quad (1)$$

can be represented as a product of electron and positron components

$$\Xi(\mathbf{r}_1^{(e)}, \dots, \mathbf{r}_N^{(e)}, \mathbf{r}_1^{(p)}, \dots, \mathbf{r}_N^{(p)}) = \Psi^{(e)}(\mathbf{r}_1^{(e)}, \dots, \mathbf{r}_N^{(e)}) \Phi^{(p)}(\mathbf{r}_1^{(p)}, \dots, \mathbf{r}_N^{(p)}). \quad (2)$$

In this case, the following normalization condition is fulfilled:

$$\langle \Psi^{(e)} | \Psi^{(e)} \rangle = \langle \Phi^{(p)} | \Phi^{(p)} \rangle = 1.$$

Here and more throughout the text, indexes i, j correspond to occupied and m, n – non-occupied (virtual) one-particle states, respectively; $\mathbf{r}^{(e)}, \mathbf{r}^{(p)}$ – electron- and positron-position vectors.

Within Hartree-Fock approximation, the total electron and positron wave functions are presented by Slater determinants:

$$\Psi^{(e)} = \frac{1}{\sqrt{N!}} \sum_P (-1)^P \varphi_1(\mathbf{r}_1^{(e)}) \times \quad (3)$$

$$\begin{aligned} & \times \varphi_2(\mathbf{r}_2^{(e)}) \cdot \dots \cdot \varphi_N(\mathbf{r}_N^{(e)}); \\ \Phi^{(p)} = & \frac{1}{\sqrt{N!}} \sum_p (-1)^p \phi_1(\mathbf{r}_1^{(p)}) \times \\ & \times \phi_2(\mathbf{r}_2^{(p)}) \cdot \dots \cdot \phi_N(\mathbf{r}_N^{(p)}), \end{aligned} \quad (3)$$

composed of one-particle wave functions obtained by solving the system of Hartree-Fock equations:

$$\begin{aligned} & -\frac{\Delta_i}{2} \varphi_i(\mathbf{r}) + (U_H^{(e)}(\mathbf{r}) - U_H^{(p)}(\mathbf{r}) + \\ & + U_{ex}^{(e)}(\mathbf{r})) \varphi_i(\mathbf{r}) = \varepsilon_i^{(e)} \varphi_i(\mathbf{r}); \\ & -\frac{\Delta_a}{2} \phi_a(\mathbf{r}) + (U_H^{(p)}(\mathbf{r}) - U_H^{(e)}(\mathbf{r}) + \\ & + U_{ex}^{(p)}(\mathbf{r})) \phi_a(\mathbf{r}) = \varepsilon_a^{(p)} \phi_a(\mathbf{r}). \end{aligned} \quad (4)$$

Here $U_H^{(e)}$ and $U_H^{(p)}$ are the corresponding Hartree's potentials defined as

$$\begin{aligned} U_H^{(e)}(\mathbf{r}) &= \int \frac{\rho^{(e)}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'; \\ U_H^{(p)}(\mathbf{r}) &= \int \frac{\rho^{(p)}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}', \end{aligned} \quad (5)$$

where

$$\begin{aligned} \rho^{(e)}(\mathbf{r}) &= 2 \sum_i^{occ} \varphi_i^*(\mathbf{r}) \varphi_i(\mathbf{r}); \\ \rho^{(p)}(\mathbf{r}) &= 2 \sum_a^{occ} \phi_a^*(\mathbf{r}) \phi_a(\mathbf{r}) \end{aligned} \quad (6)$$

are the densities of electron and positron subsystems. The non-local exchange potentials $U_{ex}^{(e)}$, $U_{ex}^{(p)}$ in (4) can be written in the following form:

$$\begin{aligned} U_{ex}^{(e)}(\mathbf{r}) \varphi_i(\mathbf{r}) &= - \sum_j^{occ} \int \frac{\varphi_j^*(\mathbf{r}') \varphi_i(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \varphi_j(\mathbf{r}); \\ U_{ex}^{(p)}(\mathbf{r}) \phi_a(\mathbf{r}) &= - \sum_b^{occ} \int \frac{\phi_b^*(\mathbf{r}') \phi_a(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \phi_b(\mathbf{r}). \end{aligned} \quad (7)$$

Note, that in the case of central symmetry one-particle wave function can be composed as a product of radial, angular and spin components:

$$\varphi(\mathbf{r}, \sigma) = \frac{P_{nl}(r)}{r} Y_{lm}(\theta, \varphi) \chi_\mu(\sigma) \quad (8)$$

and characterized by well-known quantum number set n, l, m, μ [16].

It was shown [15, 17] that in the case of equal masses of interacting particles $m_e = m_p = m$ the minimal value of the total energy corresponds to the equality of local Hartree potentials

$$U_H^{(e)}(\mathbf{r}) = U_H^{(p)}(\mathbf{r}) \quad (9)$$

and the local electrical neutrality of the system

$$\rho^{(e)}(\mathbf{r}) - \rho^{(p)}(\mathbf{r}) = 0. \quad (10)$$

So, Hartree-Fock equations are transformed to the following form [14, 15]:

$$\begin{aligned} & -\frac{\nabla^2}{2} \varphi_i(\mathbf{r}) - \\ & - \sum_j^{occ} \int \frac{\varphi_j^*(\mathbf{r}') \varphi_i(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \varphi_j(\mathbf{r}) = \varepsilon_i \varphi_i(\mathbf{r}). \end{aligned} \quad (11)$$

and the interaction between the particles is only presented by the exchange term (7). Note that the equations (11) are equally suitable for both electron and positron subsystems.

The total energy of the droplet E_{tot} consisting of N electron-positron pairs is determined by the total energies of electron E_e and positron E_p subsystems and the energy of interaction between them E_{e-p} . It can be shown that within the HF approach, the total energy of the droplet is represented by the following expression [12]:

$$E_{tot} = 2 \left(\sum_{i=1}^{occ} \varepsilon_i + E_{exch} \right). \quad (12)$$

Here, the values ε_i are one-particle energies of occupied states which are the eigenvalues of HF equation system (11), E_{exch} is the exchange interaction energy

$$E_{exch} = \frac{1}{2} \sum_{i,j=1}^{occ} \langle ij | V | ji \rangle, \quad (13)$$

where $\langle \alpha\beta | V | \gamma\delta \rangle$ is the Coulomb matrix element:

$$\begin{aligned} \langle \alpha\beta | V | \gamma\delta \rangle &= \delta_{\sigma_\alpha \sigma_\gamma} \delta_{\sigma_\beta \sigma_\delta} \times \\ & \times \iint d\mathbf{r} d\mathbf{r}' \frac{\varphi_\alpha^*(\mathbf{r}) \varphi_\beta^*(\mathbf{r}') \varphi_\gamma(\mathbf{r}) \varphi_\delta(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}. \end{aligned} \quad (14)$$

The indexes $\sigma_\alpha, \sigma_\beta, \sigma_\gamma, \sigma_\delta$ correspond to the spin projections of the one-particle states.

Using the HF approximation, one should understand that this approximation underestimates the cluster total energy as soon as it

Table 1

Total energies per pair for electron-positron clusters with different numbers N of pairs obtained within the HF [12] and LDA [11] approaches

N	$E_{tot}^{(HF)}/N$	$E_{tot}^{(LDA)}/N$
	eV	
2	-2.96	-5.12
8	-2.76	-5.44
18	-2.70	-5.56
20	-2.68	-5.60
34	-2.68	-5.64
40	-2.64	-5.72
58	-2.66	-5.70
92	-2.62	-5.60
106	-2.60	-5.58

neglects the many-electron correlations, in particular the polarization interaction [18]. Indeed, the calculations performed within the LDA framework, that effectively takes into account the static part of many-particle correlations [11], lead to essential reduction of the total energy in comparison with the HF results [12]. The corresponding comparison is shown in Table 1.

On the other hand, starting from the HF approach, one can take into account the

polarization interaction using Möller – Plesset approximation (MP2) [19], i. e. the second order of the perturbation theory. The corresponding correction of the total energy can be composed of three components

$$\Delta E^{(MP2)} = E_{e-e}^{(MP2)} + E_{p-p}^{(MP2)} + E_{e-p}^{(MP2)} \quad (15)$$

where

$$E_{e-e}^{(MP2)} = E_{p-p}^{(MP2)} = -\frac{1}{2} \sum_{ij,mn} \left(\frac{2 \langle ij | V | mn \rangle \langle mn | V | ij \rangle}{\varepsilon_m + \varepsilon_n - \varepsilon_i - \varepsilon_j} - \frac{\langle ij | V | nm \rangle \langle mn | V | ij \rangle}{\varepsilon_m + \varepsilon_n - \varepsilon_i - \varepsilon_j} \right); \quad (16)$$

$$E_{e-p}^{(MP2)} = -\sum_{ij,mn} \frac{2 \langle ij | V | mn \rangle \langle mn | V | ij \rangle}{\varepsilon_m + \varepsilon_n - \varepsilon_i - \varepsilon_j}. \quad (17)$$

The factor «2» in the formulae (16), (17) results from summation over spin variables in matrix elements (14). Substituting (16), (17) into (15) one obtains the correction to the total energy of the droplet within MP2 approximation:

$$\Delta E^{(MP2)} = -\sum_{ij,mn} \left(\frac{4 \langle ij | V | mn \rangle \langle mn | V | ij \rangle}{\varepsilon_m + \varepsilon_n - \varepsilon_i - \varepsilon_j} - \frac{\langle ij | V | nm \rangle \langle mn | V | ij \rangle}{\varepsilon_m + \varepsilon_n - \varepsilon_i - \varepsilon_j} \right). \quad (18)$$

The results of the calculations of the total energy per electron-positron pair

Table 2

The total energies per pair for electron-positron clusters with different number N of pairs obtained within the HF approach [12], and with accounting for the correlations within Möller-Plesset approximation and RPAE (present work)

N	$E_{tot}^{(HF)}/N$	$(E_{tot}^{(HF)} + \Delta E^{(MP2)})/N$	$(E_{tot}^{(HF)} + \Delta E^{(RPAE)})/N$
	eV		
2	-2.96	-4.63	-6.20
8	-2.76	-5.06	-7.22
18	-2.70	-5.26	-7.50
20	-2.68	-5.36	-7.68
34	-2.68	-5.38	-7.62
40	-2.64	-5.50	-7.94
58	-2.66	-5.38	-7.48
92	-2.62	-5.40	-7.13
106	-2.60	-5.42	-7.14

$(E_{tot}^{(HF)} + \Delta E^{(MP2)}) / N$ for the series of clusters with closed shells are presented in Table 2. Accounting for polarization interaction within the MP2 decreases the energies per pair and brings the obtained values closer to the LDA results.

However, as it was shown earlier in [15, 17], the Möller – Plesset approximation is still not able to include all correlation effects in the strongly interacting many-body systems. For the proper outline, it is necessary to consider the higher orders of perturbation theory. For this purpose, we use the Random Phase Approximation with Exchange (RPAE) which takes into account so-called «ring» Feynman diagrams of infinite order [18].

The correlated excited state can be represented within the RPAE as

$$|\Phi_v\rangle = \sum_{im} (X_{im}^v \hat{a}_m^+ \hat{a}_i - Y_{ia}^v \hat{a}_i^+ \hat{a}_m) |\Phi_0\rangle, \quad (19)$$

where $\Phi_0 = \frac{1}{\sqrt{N!}} \text{Det}\{\varphi_i(\mathbf{r}_j)\}$ is the ground state wave function within the HF approximation, \hat{a}_α^+ и \hat{a}_β are one-particle Fermi creation and annihilation operators. The coefficients X_{mi}^v and Y_{mi}^v called the «time forward» and «time reverse» amplitudes, respectively, are responsible for contributing the determinants $\hat{a}_m^+ \hat{a}_i |0\rangle$ and $\hat{a}_i^+ \hat{a}_m |0\rangle$ into the state (19). Note, that due to the local electrical neutrality of an electron-positron droplet [10 – 12], the expression (19) equally describes both the wave functions of electron and positron subsystems. It was shown [17] that the components of \mathbf{X}^v and \mathbf{Y}^v vectors can be found by solving the following RPAE matrix equation:

$$\begin{pmatrix} \mathbf{A}^{(e)} & \mathbf{B}^{(e)} & \mathbf{C} & \mathbf{D} \\ \mathbf{B}^{(e)*} & \mathbf{A}^{(e)*} & \mathbf{D}^* & \mathbf{C}^* \\ \mathbf{C} & \mathbf{D} & \mathbf{A}^{(p)} & \mathbf{B}^{(p)} \\ \mathbf{D}^* & \mathbf{C}^* & \mathbf{B}^{(p)*} & \mathbf{A}^{(p)*} \end{pmatrix} \begin{pmatrix} \mathbf{X}^{(e)} \\ \mathbf{Y}^{(e)} \\ \mathbf{X}^{(p)} \\ \mathbf{Y}^{(p)} \end{pmatrix} = \Omega \begin{pmatrix} \mathbf{X}^{(e)} \\ -\mathbf{Y}^{(e)} \\ \mathbf{X}^{(p)} \\ -\mathbf{Y}^{(p)} \end{pmatrix}, \quad (20)$$

which has $2(N_{eh} + N_{ph})$ independent solutions, where $N_{eh} = N_{ph}$ is the number of one-particle

electron-hole and positron-hole pairs. The elements of Hermitian matrixes \mathbf{A} and \mathbf{B} , including electron-electron (and positron-positron) interaction only, are described as

$$\begin{aligned} A_{im,jn}^{(e)} &= \delta_{ij} \delta_{mn} \omega_{im}^{(e)} + \langle in || mj \rangle; \\ B_{im,jn}^{(e)} &= \langle ij || mn \rangle; \\ A_{as,bt}^{(p)} &= \delta_{ab} \delta_{st} \omega_{as}^{(p)} + \langle at || sb \rangle; \\ B_{as,bt}^{(p)} &= \langle ab || st \rangle, \end{aligned} \quad (21)$$

where indexes i, j, a, b correspond to the occupied and m, n, s, t – to virtual one-particle states.

Each matrix element $\langle \alpha\beta || \gamma\delta \rangle = 2 \langle \alpha\beta | V | \gamma\delta \rangle - \langle \alpha\beta | V | \delta\gamma \rangle$ consists of «direct» and «exchange» parts defined by (14), and the factor «2» in the direct part arises from summation over spin projections.

The matrixes \mathbf{C} and \mathbf{D} correspond to the electron-positron interaction and therefore include direct interaction part only:

$$\begin{aligned} C_{im,as} &= -2 \langle is | V | ma \rangle, \\ D_{im,as} &= -2 \langle ia | V | ms \rangle. \end{aligned} \quad (22)$$

In case when one of the subsystems (for example, positrons) is «frozen» with respect to small deviation, i. e. $\delta\rho^{(p)}(\mathbf{r}) = 0$, the equation (20) reduces to the form of standard RPAE equation [18]:

$$\begin{pmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B}^* & \mathbf{A}^* \end{pmatrix} \begin{pmatrix} \mathbf{X} \\ \mathbf{Y} \end{pmatrix} = \Omega \begin{pmatrix} \mathbf{X} \\ -\mathbf{Y} \end{pmatrix}. \quad (23)$$

The elements of the eigenvectors of matrix of equation (20) satisfy the following orthonormality conditions:

$$\begin{aligned} \sum_{mi} (X_{mi}^v X_{mi}^\mu - Y_{mi}^\mu Y_{mi}^v) &= \delta_{v\mu}; \\ \sum_v (X_{mi}^v X_{nj}^v - Y_{mi}^{v*} Y_{nj}^v) &= \delta_{mn} \delta_{ij}, \end{aligned} \quad (24)$$

where index v corresponds to an eigenvector with eigenvalue Ω_v that equals to the transition frequency between the ground state and v -th excited one.

Due to the symmetry of equation (20), there are two solutions with eigenvalues Ω_v and $-\Omega_v$ and eigenvectors $(\mathbf{Y}_v^{(e)}, \mathbf{X}_v^{(e)}, \mathbf{Y}_v^{(p)}, \mathbf{X}_v^{(p)})$ and $(\mathbf{X}_v^{(e)}, \mathbf{Y}_v^{(e)}, \mathbf{X}_v^{(p)}, \mathbf{Y}_v^{(p)})$, respectively. This means that one should consider only $(N_{eh} + N_{ph})$ solutions with positive eigenvalues.

Besides, if the masses of particles are equal ($m_e = m_p = m$), then, due to the symmetry between two subsystems

$$\begin{aligned}\mathbf{A}^{(e)} &= \mathbf{A}^{(p)}, \\ \mathbf{B}^{(e)} &= \mathbf{B}^{(p)},\end{aligned}\quad (25)$$

one can distinguish two following types of the solutions of equation (20): the symmetric modes, that match the conditions $\mathbf{X}^{(e)} = \mathbf{X}^{(p)}$, $\mathbf{Y}^{(e)} = \mathbf{Y}^{(p)}$, and the antisymmetric ones with the relations $\mathbf{X}^{(e)} = -\mathbf{X}^{(p)}$, $\mathbf{Y}^{(e)} = -\mathbf{Y}^{(p)}$. The solutions of the first type correspond to «acoustic» type of oscillations of the particle density, which retains the local electrical neutrality of the whole system. Thus, in the calculations of optical characteristics of electron-positron droplets, we should take into account only antisymmetric (dipole or «optical») modes. This allows us to transform the equation (20) into the equivalent form with matrix size $2N_{eh} \times 2N_{eh}$ with N_{eh} independent solutions [15, 17]:

$$\begin{pmatrix} \tilde{\mathbf{A}} & \tilde{\mathbf{B}} \\ \tilde{\mathbf{B}}^* & \tilde{\mathbf{A}}^* \end{pmatrix} \begin{pmatrix} \mathbf{X} \\ \mathbf{Y} \end{pmatrix} = \Omega^{(RPAE)} \begin{pmatrix} \mathbf{X} \\ -\mathbf{Y} \end{pmatrix}, \quad (26)$$

where

$$\begin{aligned}\tilde{A}_{im,jn} &= A_{im,jn} + C_{im,jn} = \\ &= \omega_{im} \delta_{ij} \delta_{mn} + 4 \langle in | V | mj \rangle - \langle in | V | jm \rangle, \\ \tilde{B}_{im,jn} &= B_{im,jn} + D_{im,jn} = \\ &= 4 \langle ij | V | mn \rangle - \langle ij | V | nm \rangle.\end{aligned}\quad (27)$$

Note, that the orthonormality conditions (24) for eigenvectors of equation (26) remain intact.

The coefficients Y_{mi}^v obtained by solving the equation (26) assess the contribution of the correlations to the ground HF state. Finally, the correlation correction to the HF ground state energy within RPAE can be written as [21]

$$\Delta E^{(RPAE)} = - \sum_{im,v} \Omega_v^{(RPAE)} |Y_{im}^v|^2, \quad (28)$$

where the summation is performed over all excited states $|\Phi_v\rangle$, the energies $\Omega_v^{(RPAE)}$ are the eigenvalues of equation (26).

Note that the same result can be obtained within Tamm – Dancoff approximation [21]

$$\Delta E^{(RPAE)} = - \frac{1}{2} \sum_v (\Omega_v^{(RPAE)} - \Omega_v^{(TDA)}) \quad (29)$$

where $\Omega_v^{(TDA)}$ are the eigenfrequencies of the corresponding Tamm – Dancoff equation

$$\tilde{\mathbf{A}}\mathbf{X} = \Omega^{(TDA)}\mathbf{X}. \quad (30)$$

Besides the correlation contribution to the ground state energy, the correlations significantly affect the optical properties of many-particle system. Indeed, the static dipole polarizability of an electrically neutral electron-positron droplet can be calculated as [15, 17]

$$\alpha^{(e+p)} = \sum_v \frac{f_v}{\mu \Omega_v^2}. \quad (31)$$

Here the summation is over all excited states of the system, μ is the reduced mass of the electron-positron pair

$$\mu = \frac{m_e m_p}{m_e + m_p} = \frac{m}{2},$$

f_v are the dipole transition oscillator strengths between the ground state and the v -th excited many-particle state with frequency Ω_v . The oscillator strengths are defined by the following expression:

$$f_v = \frac{4}{3} \mu \Omega_v D_v^2. \quad (32)$$

They satisfy the sum rule $\sum_v f_v = N$ [18, 21]. The transition matrix elements D_v are calculated by summation over all the one-particle excited states:

$$D_v = \sum_{im} (X_{im}^v d_{im} + Y_{im}^v d_{mi}), \quad (33)$$

where d_{im} are the single-particle dipole amplitudes in the length gauge [18]:

$$d_{im} = \int P_{n_i l_i}(r) r P_{n_m l_m}(r) dr, \dots, l_m = l_i \pm 1. \quad (34)$$

III. Results of Calculations

In order to solve the equations (20) and (26) it is necessary to have a complete basis of the one-particle wave functions (8) including both discrete and continuum spectra states. To perform the numerical calculations, we use replacing the true virtual one-particle states by the discrete spectrum of pseudostates. This approach is based on the representation of the radial wavefunctions (8) as a linear combination of the piecewise continuous polynomials defined on a sufficiently large radial interval $[0, R_{\max}]$, so-called B-splines [22]. The new rede-

finned functions $P_{nl}(r)$ satisfy the boundary conditions $P_{nl}(0) = P_{nl}(R_{\max}) = 0$, i. e. the system is supposed to be placed in the spherical potential well with the walls of infinite height. This approach allows us to carry out summation over a finite number of discrete pseudostates instead of integrating over continuous part of the spectrum.

In our calculations we use 50 B -splines of the order 7 for each orbital quantum number l , and implement the summation over transferred angular momenta up to $l = 10$. The partial contributions of the terms with different transferred angular momenta to the correlational part of the energy are shown in Fig. 1 for the electron-positron clusters of different sizes. One can see that for small clusters the main contribution comes from the dipole term while the contributions with larger transferred momenta rapidly decrease. At the same time, for clusters with $N > 20$ the terms with larger transferred momenta play more significant role and their contributions decrease more slowly with l .

The results of the calculations of the total energies per electron-positron pair for the clusters of different size are presented in Table 2 and Fig. 2. For comparison, we present the results obtained within the HF approximation, the LDA, the second order

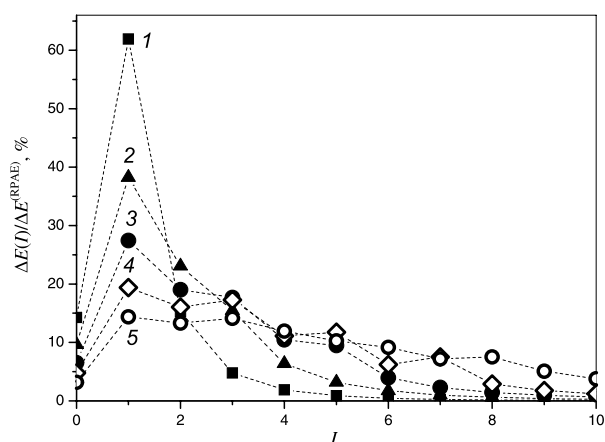


Fig. 1. The partial contributions to the correlation energy of electron-positron clusters as a function of transferred angular momentum for the systems with different number of electron-positron pairs: $N = 2$ (1), $N = 8$ (2), $N = 20$ (3), $N = 40$ (4), $N = 92$ (5)

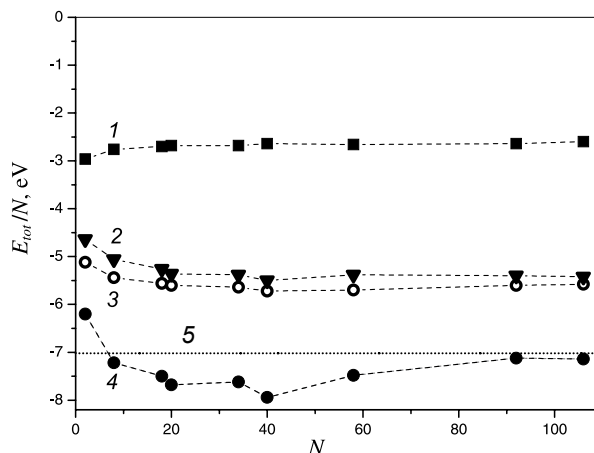


Fig. 2. The total energies of clusters per electron-positron pair as a function of cluster size obtained within the different theoretical approaches: 1 – HF [12], 2 – LDA [11], 3 – MP2, 4 – RPAE; 5 – the result of *ab initio* calculations of dipositronium molecule [4]

Möller – Plesset approximation (MP2) and the RPAE as well.

As it can be seen from Table 2, as well as from Fig. 2, the closed-shell clusters containing 20, 34 and 40 electron-positron pairs have the lowest values of the energy. There is a conspicuous minimum of the total energy calculated with the correlation corrections, while the

Table 3

Reduced static dipole polarizabilities $\alpha^{(e+p)}/R^3$ for the clusters consisting of the different numbers of electron-positron pairs N . For comparison, there are reduced polarizabilities of electron subsystem presented here and obtained in a frozen positively charged core $\alpha^{(e)}/R^3$

N	$\alpha^{(e+p)}/R^3$	$\alpha^{(e)}/R^3$
2	1.40	1.24
8	1.58	1.39
18	1.46	1.29
20	1.54	1.38
34	1.39	1.25
40	1.50	1.37
58	1.33	1.21
92	1.30	1.20
106	1.31	1.21

energies within Hartree – Fock approximation have approximately equal values for almost all of the clusters considered (except for the case of $N = 2$). Besides, the calculations with the correlation corrections $(E_{tot}^{(HF)} + \Delta E^{(RPAE)})/N$ show that the pair energy of two electron-positron cluster is higher than half of the total energy of the molecule dipositronium (7.02 eV) obtained in the *ab initio* calculations [4], while the pair energies of other systems are visibly lower.

Thus, it can be assumed that formation of a cluster consisting of two electron-positron pairs bound by exchange-correlation interaction is energetically unfavorable in comparison with the positronium molecule the structure of which is determined by van der Waals interaction [8].

As for the larger systems, the cluster structure of an electron-positron droplet becomes more beneficial. In favor of the latter statement, the behavior of the reduced static dipole polarisability with cluster size reveals the maximum values for $N = 8 - 40$. Table 3 presents the ratios of the static dipole polarizability of electron-positron droplets calculated in RPAE, to the value of the effective cluster radius in the third degree. The effective radius is calculated by the simple formulae

$$R = \sqrt[3]{\frac{3}{4\pi \bar{n}}}, \quad (35)$$

where \bar{n} is the average concentration of electron-positron pairs in the system:

$$\bar{n} = \frac{1}{N} \int \rho_e(\mathbf{r})\rho_p(\mathbf{r})d\mathbf{r}. \quad (36)$$

For the systems of medium size $N = 8 - 40$ there is an admittedly optimal proportion of the particles in surface area, that give the main contribution to the polarization interaction, with respect to the volume area particles.

It should be noted that, as one may see from Table 3, the polarizability of mutually correlated electron-positron droplets $\alpha^{(e+p)}$ for all N is somewhat higher than the corresponding values calculated in the frozen core of one of the subsystems $\alpha^{(e)}$. It means that the correlations between the particles constituting the cluster subsystems influence not only the stability of the electron-positron droplets as a whole, but also their optical properties.

IV. Concluding Remarks

This work presents the results of the total energy calculations as well as the static polarisability of electron-positron clusters with closed shells. The calculations fulfilled show that the single-particle approximations like Hartree – Fock one and the Local Density Approximations are not able to provide a stable state of such systems. The total energies obtained within HF and LDA calculations are higher than the corresponding energies of electron-positron systems bound by van der Waals interactions.

However, using HF approximation as a zero guess, it is possible to take into account additional correlations aroused by the strong polarization interaction of electron-positron system. The calculations with accounting for the second order of perturbation theory and within the RPAE reveal the significant decrease of the total energies.

In particular, the RPAE results for the clusters with the number of pairs $N \geq 8$ are below the energy level of -7.02 eV, which is equal to the total energy per pair of positronium molecule [4]. The calculations reveal the minimum of the total energy per pair for the numbers of N in the range between 20 and 40, and this corresponds to the most stable electron-positron clusters. This minimum could be understood if one considered the behavior of reduced static dipole polarizability of electron-positron systems, which also has a visible maximum in the same size region.

The existence of stable electron-positron clusters could be investigated experimentally. These objects can be created during condensation or density collapse of electrons and positrons in a certain volume. The necessary conditions for such process can be obtained in experiments with electron and positron aligned beams of high density and of equal energy. The stable clusters can be determined by the methods of optical spectroscopy. Indeed, the relevant time corresponding to the frequency of dipole resonance is about ~ 1.5 fs [17] while the lifetime of the annihilation decay of dipositronium molecular is the five orders of magnitude larger [5]. This condition can provide one with enough life time of electron-positron cluster to study its optical properties.

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