Complex exponential basis wave functions in the Coulomb three-body problem

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A new type of basis function for variational calculations of quantum-mechanical systems with Coulomb interaction between the particles is proposed, viz. complex exponential functions of the interparticle distances. The simplicity of the calculational formulas and of the computer programs that is characteristic of exponential functions is retained when the proposed basis functions are employed; the latter, however, yield much better convergence of the variational calculations compared to the usual real exponential basis functions. This is particularly true in the case of molecule-like systems in which two heavy particles of like charge are bound by a light particle. On the basis of calculations of various three-particle Coulomb systems it is shown that a single real exponential-trigonometric basis function consisting of a pair of complex conjugate exponential functions can replace from 10 to 15 real exponential functions in calculations of atom-like systems and up to 80 such functions in calculations of molecule-like systems. Thus the proposed new basis functions can significantly broaden the scope of variational calculations of Coulomb systems without complicating the method of calculation.

The problem of quantum-mechanical systems with a small number of particles interacting according to the Coulomb law has stood in recent decades at the center of attention of a large number of authors and groups of authors. It is important for the theory of mesomolecules and mesomolecular ions, which participate in the muon catalysis of reactions of nuclear synthesis, and also in the detailed theory of atoms, molecules, and their electron-hole analogs in the solid state. At present two important directions in the approach to this problem have taken shape: an approach based on the expansion of the wave functions of the systems over the adiabatic basis,[1-3] and the direct variational method, based on the expansion of the unknown wave functions over basis functions of simple analytic form, among which we may note in particular the Hylleraas functions[4-7] and the purely exponential functions.[8-14] These approaches to a certain extent complement each other. The expansion over the adiabatic basis is the more applicable of the two in the case of heavy identically charged particles bound by a light oppositely charged particle and converges poorly in the case of light particles bound by a heavy particle. The direct variational methods are the more applicable of the two in the region where the adiabatic expansion converges poorly, but cease to be of service in the limit of infinitely heavy bound particles and a light binding particle, i.e., when the adiabatic basis of one single function gives the exact solution of the problem. This justifies the parallel development of both approaches, which in their regions of applicability (and using a wide enough basis) give precise results for the energies of various three-particle Coulomb systems, reviews of which can be found, e.g., in Refs. 2, 3, 7, and 13. At the same time, it should be noted that the adiabatic expansion method, which requires the preliminary solution of a quite complicated Schrödinger equation to determine the basis functions, is more labor-intensive than the direct variational method.

In the ground-breaking work of Delves and Kalotas,[8] dedicated to the mesomolecular ion $d^+d^+\mu^-$, was proposed a simple and practical method for variational calculations of three-particle Coulomb systems with complete account of the motion of all three particles. It is based on the use of basis functions which depend exponentially on the interparticle distances. Its unquestionable virtue is the exceptional simplicity of the calculations of the matrix elements of the energy operator, which are easily performed in perimetric coordinates and yield compact analytic formulas suitable for numerical computation. After a fifteen-year pause this method was discovered anew in Refs. 9 and 10, and after a detailed reworking of its purely computational aspects it has now become one of the main variational methods for calculating three-particle Coulomb systems; a detailed exposition of the technique of this method and the results obtained using it can be found in Refs. 11–14.

As in other variational methods, the convergence of this variational method deteriorates as one goes from atomic systems (where two light particles are bound by one heavy particle) to a system with equal particle masses, the positronium ion $e^-e^-e^+$, and from there to mesomolecular ions and, finally, to isotopic modifications of the molecular hydrogen ion H_2^+. Thus, for the mesomolecular deuterium ion $d^+d^+\mu^-$, using N exponential basis functions (with three nonlinear parameters entering into the exponents) the total energy E is reproduced with $N=15$ but only to two significant figures, with $N=35$, to three significant figures, and to six significant figures starting with $N=125$. As to the molecular ion $H_2^+(\infty)$ with fixed (infinitely heavy) nuclei, for its energy even with $N=350$ one obtains the value $E\approx -0.592010$ instead of the exact (rounded off to six significant figures) $E=-0.602634$, which corresponds to a 10% error in the binding energy of this system (see Refs. 11 and 12). The reason for the poor convergence of the variational calculations in molecules with heavy nuclei is that the set of monotonic exponential functions of the interparticle distances is poorly suited to approximate the vibrational wave function of a molecule that has a sharp maximum near the equilibrium of the nuclear shell. This feature of the variational method is particularly pronounced in the case of the

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molecular hydrogen ion with fixed nuclei, for which the vibrational function becomes a delta-function.

Because of this peculiarity of purely exponential basis functions, to obtain precise results it is necessary to use bases that contain hundreds (sometimes even thousands) of functions, which, in turn, is associated with difficulties that arise as a consequence of the nearness to zero of the determinants of the matrices which are made up from overlap integrals of the basis functions.

The goal of the present paper is to show that the capabilities of the exponential functions can be significantly extended without complicating the corresponding analytic and calculational work by using a new type of basis function: complex exponential functions of the interparticle distances.

Let us consider a three-particle system with particle charges Z_j and masses m_j, where $j = 1, 2, 3$. For definiteness we take the signs of the charges of the first two particles to be identical and opposite that of the third particle (the binding particle). We then have $Z_1Z_2 > 0$, $Z_2Z_3 > 0$, $Z_3Z_1 < 0$. Introducing as the relative coordinates the interparticle distances r_{12}, r_{23}, and r_{31}, and restricting ourselves (for the sake of simplicity) to the states of the system with zero total angular momentum, we have for the Hamiltonian of the system the following expression:

$$H=\sum_{j=1}^{3}\left[-\frac{1}{2\mu_j}\left(\frac{\partial^2}{\partial r_j^2}+\frac{2}{r_j}\frac{\partial}{\partial r_j}\right)-\frac{\cos\theta_j}{m_j}\frac{\partial^2}{\partial r_{j+1}\partial r_{j+2}}+\frac{Z_{j+1}Z_{j+2}}{r_j}\right]. \tag{1}$$

We abbreviate the notation for the interparticle distances as follows:

$$r_1=r_{23},\ r_2=r_{31},\ r_3=r_{12}. \tag{2}$$

The quantities μ_j are the reduced masses of the particle pairs:

$$\mu_j=m_{j+1}m_{j+2}/(m_{j+1}+m_{j+2}), \tag{3}$$

and θ_j denotes the angle between the line segments connecting the jth particle with the other two particles. Then

$$\cos\theta_j=(r_{j+1}^2+r_{j+2}^2-r_j^2)/(2r_{j+1}r_{j+2}). \tag{4}$$

When the index of some quantity (r_{j+k}, m_{j+k}, or Z_{j+k}) is greater than three, it must be reduced by three (i.e., $r_4 = r_1$, $r_5 = r_2$, $m_4 = m_1$, $m_5 = m_2$, $Z_4 = Z_1$, and $Z_5 = Z_2$).

The basis functions proposed here have the following form:

$$\varphi_k(r_1, r_2, r_3)=\exp\left(-\sum_{j=1}^{3}A_{kj}r_j\right), \tag{5}$$

where A_{k1}, A_{k2}, and A_{k3} are a triple of complex numbers which characterizes the basis function with index k. Here

$$A_{kj}=a_{kj}+ib_{kj},\ j=1,\ 2,\ 3, \tag{6}$$

so that each basis function is determined by six real numbers a_{k1}, a_{k2}, a_{k3}, b_{k1}, b_{k2}, and b_{k3}.

The normalization and overlap integrals of the basis functions (5), and also the matrix elements of the Hamiltonian (1) in this basis, are easily calculated in perimetric coordinates (see Appendix).

Since the wave function of the system of particles in the present case, without a magnetic field, should be real, to each basis function φ_k (5) there should correspond a complex conjugate basis function φ_k^*. Out of the basis functions φ_k and φ_k^* it is possible to construct a pair of real basis functions

$$\varphi_k^{(+)}=\frac{1}{2}(\varphi_k+\varphi_k^*)=\exp\left(-\sum_{j=1}^{3}a_{kj}r_j\right)\cos\left(\sum_{j=1}^{3}b_{kj}r_j\right), \tag{7}$$

$$\varphi_k^{(-)}=\frac{1}{2i}(\varphi_k-\varphi_k^*)=\exp\left(-\sum_{j=1}^{3}a_{kj}r_j\right)\sin\left(\sum_{j=1}^{3}b_{kj}r_j\right). \tag{8}$$

This pair of functions enters into the expansion of the unknown wave function of the system of particles together with some (determined from the variational principle) real coefficients $d_k^{(+)}$ and $d_k^{(-)}$. Therefore the contribution of the pair of functions (7), (8) to this wave function is equal to

$$d_k^{(+)}\varphi_k^{(+)}+d_k^{(-)}\varphi_k^{(-)}=C_k\exp\left(-\sum_{j=1}^{3}a_{kj}r_j\right)\sin\left(\sum_{j=1}^{3}b_{kj}r_j+\eta_k\right), \tag{9}$$

where

$$C_k=[(d_k^{(+)})^2+(d_k^{(-)})^2]^{1/2},\ \operatorname{tg}\eta_k=d_k^{(+)}/d_k^{(-)}. \tag{10}$$

Thus, the expansion of the unknown wave function of the system of particles with respect to the basis of complex exponential functions (5) containing the pair of complex conjugate functions is equivalent to expanding it over the real basis consisting of exponential-trigonometric functions of the following form:

$$f_k(r_1, r_2, r_3)=\exp\left(-\sum_{j=1}^{3}a_{kj}r_j\right)\sin\left(\sum_{j=1}^{3}b_{kj}r_j+\eta_k\right). \tag{11}$$

Each basis function f_k contains six nonlinear parameters (a_{kj} and b_{kj}) and one linear parameter, corresponding to the tangent of the angle η_k.

It is important that all of the calculations of the normalization and overlap integrals and matrix elements of the Hamiltonian in the basis of functions (11) reduce to a calculation of the analogous quantities in the basis of elementary complex exponential functions (5) via simple analytic formulas given in the Appendix, and are easily determined numerically using the capability of direct operation with complex numbers provided by the programming language FORTRAN. Therefore the calculational work in the basis of exponential-trigonometric functions (11) is as simple as using the commonly used basis of purely exponential (real) basis functions.

At the same time, thanks to its nonmonotonic dependence on the interparticle distances, the exponential-trigonometric basis functions (11) are much more flexible than the purely exponential functions.

In order to demonstrate the capabilities of these basis functions, we have carried out calculations of the ground state of a number of three-particle systems which are symmetric in the masses and charges of the first two particles ($Z_1 = Z_2 = \pm 1$, $Z_3 = \mp 1$, $m_1 = m_2$), restricting the discussion to the simple case of a variable wave function obtained from the one single basis function (11) by symmetriz-

TABLE I. Results of the variational calculations of three-particle Coulomb systems which differ in the masses of their particles, using the exponential-trigonometric test wave function (13).

System	H_2^+ (∞)	$p+p+e^-$	$\mu+\mu+e^-$	$t+t+\mu^-$	$d+d+\mu^-$
$\tilde{\alpha}$	1,0881	1,0878	1,0842	1,0629	1,0533
$\tilde{\beta}$	0,2123	0,2121	0,2105	0,2055	0,2054
$\tilde{\gamma}$	0,4420	0,4421	0,4421	0,4293	0,4215
$\tilde{\delta}$	−0,0508	−0,0510	−0,0531	−0,0624	−0,0655
$\tilde{\varepsilon}$	−0,0213	−0,0212	−0,0202	−0,0161	−0,0143
$\tilde{\zeta}$	0,7220	0,7183	0,6912	0,5612	0,5129
tg η	−0,2474	−0,2442	−0,2200	−0,1020	−0,0595
$-E_{var}$	0,57433	0,57376	0,56940	111,853	109,017
$-E_{exact}$	0,60263 [15]	0,59714 [15]	0,58512 [16]	112,973 [12]	109,817 [12]
$\Delta E/E$, %	4,7	3,9	2,7	1,0	0,7

System	$p+p+\mu^-$	$e^-e^-e^+$	$\mu+e^-e^-$	$p+e^-e^-$	H^- (∞)
$\tilde{\alpha}$	1,0314	0,9421	0,9536	0,9554	0,9560
$\tilde{\beta}$	0,2086	0,2842	0,4444	0,4470	0,4475
$\tilde{\gamma}$	0,4000	0,2542	0,0405	0,0359	0,0353
$\tilde{\delta}$	−0,0700	−0,0408	−0,0600	−0,0638	−0,0640
$\tilde{\varepsilon}$	−0,0107	0,0023	0,0027	0,0031	0,0032
$\tilde{\zeta}$	0,4155	0,0892	0,0760	0,0800	0,0804
tg η	0,0221	0,1107	0,2531	0,2714	0,2736
$-E_{var}$	101,801	0,26155	0,52436	0,52675	0,52705
$-E_{exact}$	102,224 [12]	0,26200 [12]	0,52506 [12]	0,52745 [17]	0,52775 [18]
$\Delta E/E$, %	0,41	0,17	0,13	0,13	0,13

ing it with respect to the coordinates of the identical particles 1 and 2:

$$\Psi = f_1(r_1, r_2, r_3) + f_1(r_2, r_1, r_3). \quad (12)$$

In the usual notation of the interparticle distances this wave function can be written in the form

$$\Psi = \exp(-\gamma r_{12})\,[\exp(-\alpha r_{13} - \beta r_{23})\sin(\delta r_{13} + \varepsilon r_{23} + \zeta r_{12} + \eta) + \exp(-\beta r_{13} - \alpha r_{23})\sin(\varepsilon r_{13} + \delta r_{23} + \zeta r_{12} + \eta)\,]. \quad (13)$$

In place of the subscripted parameters a_{1j} and b_{1j} we have introduced here more easily distinguished symbols.

The results of the calculations of ten three-particle systems, differing in the masses of the particles, carried out by varying the six nonlinear parameters (α, β, γ, δ, ε, and ζ) and one linear parameter (tan η) entering into the test wave function (13), are presented in Table I.

In the first seven rows of this table are given the optimal values of the variational parameters, found by minimizing the mean value of the energy (by the method of steepest descent). In the next three rows are given the variational values of the energy, the exact values of the energy of the investigated systems, and the relative error of the variational value of the energy. This error is maximal for the molecular hydrogen ion with infinitely heavy nuclei (4.7%), and falls off monotonically as a function of the ratio of the mass of the binding particle to the masses of the bound particles, and for the negative ion of the hydrogen atom stands at 0.13%. Comparison with the variational calculations of other authors shows that one exponential-trigonometric test wave function (13) in the case of the molecular hydrogen ion provides the same accuracy as 80 exponential basis functions,[12] and, in the case of the mesomolecular ion $d^+d^+\mu^-$, 23 exponential basis functions, but in the case of the H^- ion, ten Hylleraas basis functions.[11,19]

These results, obtained with one single basis function (13), provide convincing proof of the effectiveness of the exponential-trigonometric basis (11) proposed here, constructed from elementary complex exponential functions (5). These basis functions make it possible to substantially improve the modeling of the dependence of the wave functions of Coulomb systems on the interparticle distances in comparison with real exponential basis functions and to improve the convergence of the variational calculations by one to one and a half orders of magnitude.

By way of a second illustration we present calculations of the energies of the ground state of the helium atom and helium-like ions, the results of which are shown in Table II. In this case $Z_1 = Z_2 = -1$, $m_1 = m_2 = 1$, $m_3 = \infty$, and the charge of the third particle $Z_3 = Z$ runs through the values $+1$ to $+10$. The first column of Table II corresponds to calculations with a purely exponential test function of the form

$$\Psi = \exp(-\gamma r_{12})\,[\exp(-\alpha r_{13} - \beta r_{23}) + \exp(-\beta r_{13} - \alpha r_{23})], \quad (14)$$

the second column corresponds to calculations with the exponential-trigonometric test function of the form (13), the third column presents results of calculations with a 15-term Hylleraas basis,[19] and in the final column, the corresponding rounded-off results of exact calculations.[18] From this table it is clear that the compact exponential-trigonometric test function (13) in the given case is almost as accurate as the calculations of the energy in the 15-term Hylleraas basis and substantially more accurate than the results obtained with the purely exponential test wave function (14).

Let us discuss briefly the values of the variational parameters found by minimizing the mean value of the energy for systems with different ratios of the masses of the light and heavy particles, shown in Table I. Since for each of these systems the natural unit of length is determined by the re-

TABLE II. Absolute values of the ground state energy of the helium atom and helium-like ions, calculated with various variational test wave functions.

Nulear charge Z	Purely exponential function (14)	Exponential-trigonometric function (13)	15-term wave function[19]	Precise calulation[18,20]
1	0,52387	0,52705	0,52718	0,52775
2	2,89953	2,90317	2,90319	2,90372
3	7,27571	7,27929	7,27939	7,27991
4	13,65137	13,65487	13,65505	13,65557
5	22,02677	22,03023	–	22,03097
6	32,40205	32,40550	32,40550	32,40625
7	44,77725	44,78064	–	44,78145
8	59,15241	59,15575	59,15609	59,15661
9	75,52753	75,53085	–	75,53171
10	93,90262	93,90592	93,90630	93,90681

duced mass of the bound and binding particles, in place of the values of the quantities $\alpha, \beta, \gamma, \delta, \varepsilon$, and ζ, Table I gives the values of the quantities $\tilde{\alpha}, \tilde{\beta}, \tilde{\gamma}, \tilde{\delta}, \tilde{\varepsilon}$, and $\tilde{\zeta}$ defined by the formulas $\tilde{\alpha} = \alpha/\mu, \tilde{\beta} = \beta/\mu, \tilde{\gamma} = \gamma/\mu$, etc., where

$$\mu = m_1 m_3/(m_1+m_3).$$

The parameters $\tilde{\alpha}, \tilde{\beta}, \tilde{\gamma}$, and $\tilde{\zeta}$ are positive for all the systems considered here and the value of $\tilde{\delta}$ is negative for all these systems, while the quantity $\tilde{\varepsilon}$ is negative for the molecular hydrogen ion and grows as one moves toward the atom-like systems, passing through zero between the positronium ion and the mesomolecular ion $p^+p^+\mu^-$. The closeness of the absolute values of the parameters $\tilde{\delta}$ and $\tilde{\varepsilon}$ to zero means that the trigonometric correction to the exponential dependence of the wave function (13) on the distance between the oppositely charged particles r_{13} and r_{23} is relatively small. At the same time, the larger values of the parameter $\tilde{\zeta}$ (particularly for the molecule-like systems) points to the important role of such a trigonometric correction to the dependence of the test-wave function on the distance r_{12} between the like-charged particles. Of the three purely exponential parameters of the test wave function $(\tilde{\alpha}, \beta, \gamma)$, as one goes from the molecule-like systems to the atom-like systems the parameter $\tilde{\gamma}$ falls off, while the parameters $\tilde{\alpha}$ and $\tilde{\beta}$ fall off gently at first, have shallow minima in the vicinity of the positronium ion and the mesomolecular ions, respectively, and then grow.

The positive sign of the parameter $\tilde{\gamma}$, which corresponds to attraction between the like-charged particles, deserves attention. This effect of the parameter $\tilde{\gamma}$ is cancelled out by the dependence of the argument of the trigonometric function on the quantity ζr_{12}, leading (on the average) to a correct account of the correlation in the motion of particles 1 and 2. Precisely such a distribution of "roles" between the exponential and trigonometric parts of the test wave function (13) is the source of its flexibility, allowing it to compete with a linear combination of 10–80 purely exponential functions.

The quantity $\tan\eta = d^{(+)}/d^{(-)}$, which being the ratio of the contribution of the "cosinusoidal" function (7) to that of the "sinusoidal" function (8) to the test wave function (13), grows monotonically as one goes from the molecular hydrogen ion $H_2^+(\infty)$ to the atomic ion H^-, in the process passing through zero in the region between the mesomolecular ions $d^+d^+\mu^-$ and $p^+p^+\mu^-$. Here the absolute value of $\tan\eta$ has almost identical values for the ions $H_2^+(\infty)$ and $H^-(\infty)$.

Let us dwell now on the problem of the nodes of the wave function (13) that represent a formal insufficiency of this function (in the exact eigenfunction of the ground state there are no nodes). Taking into account the numerical values of the variational parameters from Table I, one can convince oneself that these nodes are located in the region of configuration space that is of little significance for the energy calculations: in these regions the exponentially decaying factor is extremely small and, for the H^- ion for example, is of order 10^{-3}.

The results obtained here show that the use of complex exponential basis functions (or the exponential-trigonometric basis functions corresponding to them) makes it possible with a minimum of effort to substantially broaden the possibilities of variational calculations of three-particle Coulomb systems, to improve their convergence, and to decrease the number of basis functions needed to achieve a given accuracy.

The transition from the case considered here of one exponential-trigonometric basis function to a superposition of such functions does not introduce any difficulties and is easily realized with the use of generally available standard programs for calculating the eigenvalues and eigenvectors of the matrices. Simultaneously, the problem of the nodes is eliminated (in analogy with the possibility of representing a node-free function by a truncated Fourier series).

APPENDIX. CALCULATION OF THE MATRIX ELEMENTS OF THE HAMILTONIAN AND THE OVERLAP INTEGRALS WITH COMPLEX EXPONENTIAL BASIS FUNCTIONS

Operating with the Hamiltonian (1) on the basis function (5), we have

$$H\varphi_k = \sum_{j=1}^{3}\left[-\frac{1}{2\mu_j}\left(A_{kj}^2 - \frac{2A_{kj}}{r_j}\right) + \frac{Z_{j+1}Z_{j+2}}{r_j} - \frac{A_{k,j+1}A_{k,j+2}}{2m_j}\cos\theta_j\right]\varphi_k. \tag{A1}$$

Hence it is obvious that calculations of the matrix elements of the Hamiltonian reduce to the calculation of the matrix elements of the operators $1/r_j$ and $\cos\theta_j$ and the overlap integrals. We denote by φ_j a basis function of the form (5) with parameters A_{j1}, A_{j2}, and A_{j3}. The required integrals are elementary to calculate in perimetric coordinates. We have

$$\langle\varphi_l|\varphi_k\rangle=\frac{2}{(C_1C_2C_3)^2}\left(1+\frac{C_2+C_3}{C_1}+\frac{C_3+C_1}{C_2}+\frac{C_1+C_2}{C_3}\right), \tag{A2}$$

$$\left\langle\varphi_l\left|\frac{1}{r_1}\right|\varphi_k\right\rangle=\frac{1}{(C_1C_2C_3)^2}\left(C_1+C_2+C_3+\frac{2C_2C_3}{C_1}\right), \tag{A3}$$

$$\langle\varphi_l|\cos\theta_1|\varphi_k\rangle=\frac{2}{(C_1C_2C_3)^2}\left(1+\frac{C_2+C_3}{C_1}+\frac{C_3-C_1}{C_2}+\frac{C_2-C_1}{C_3}\right). \tag{A4}$$

The complex quantities C_1, C_2, and C_3 are given by the formula

$$C_j=A_{k,j+1}+A_{k,j+2}+A^{*}_{l,j+1}+A^{*}_{l,j+2}. \tag{A5}$$

The matrix elements of the operators $1/r_2$, $1/r_3$, and $\cos\theta_3$ are obtained from formulas (A3) and (A4) by cyclic permutation of the indices. As in the main text, the condition for lowering the indices $j+1$ and $j+2$ by three is used as needed.

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