PHYSICS OF ELEMENTARY PARTICLES AND ATOMIC NUCLEI. THEORY

Energy Levels of a Helium Atom

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Abstract—The nonrelativistic ionization energy levels of a helium atom are calculated for S, P, and D states. The calculations are based on the variational method of "exponential" expansion. The convergence of the calculated energy levels is studied as a function of the number of basis functions (N). This allows us to claim that the obtained energy values (including the values for the states with a nonzero angular momentum) are accurate to 20 significant digits.

INTRODUCTION

The quantum problem of three bodies with Coulomb interaction is one of the most notable nonintegrable problems in quantum mechanics. At the same time, extremely accurate numerical solutions for the problem of bound states for a system of three particles may be obtained with modern computers. For example, the nonrelativistic energy of the ground state of helium with a nucleus of an infinite mass is now known accurately to 46 significant digits [1].

In the present study, a version of the variational method (the so-called "exponential" expansion) that renders it possible to numerically solve the quantum Coulomb three-body problem with a very high accuracy and is applicable to, among other things, states with a nonzero angular momentum is considered. This method is used to calculate the nonrelativistic ionization energies of a helium atom for S, P, and D states. It is shown that the developed method is an efficient and flexible instrument for investigating Coulomb systems. An analysis of convergence proves that the method is highly accurate and demonstrates that nonrelativistic energy values accurate to 20 significant digits may be obtained easily.

The development of such high-precision methods is also important for the reason that it may help solve a wide variety of physical problems that are of interest in practice. For example, atoms of antiprotonic helium $He^+\bar{p}$ are studied in physics of exotic atoms and molecules [2]. One of the electrons of helium is replaced by an antiproton in this atom. Under certain conditions, this antiproton forms metastable states with a lifetime of several microseconds. Not only is this a record lifetime for an antiparticle within a standard (real) medium, but also an astronomical time by the standards of atomic physics, where the lifetime of the $2P$ state of a hydrogen atom is only 10 ns. This allows one to use precision laser spectroscopy to probe the structure of the spectrum of an antiprotonic helium atom.

Another very important aspect, namely, the cross impact of atomic and nuclear physics [3], in the determination of statistical parameters of nuclei should be noted. For example, the accuracy of the mean-square helium charge radius that is determined experimentally from electron–nuclei scattering is about $1-3\%$. At the same time, the experimental determination of the charge radius of ^4He by muonic atom spectroscopy allows one to reduce the error in the value of this parameter by more than an order of magnitude.

The paper is structured as follows. The application of the variational method to the stationary Schrödinger problem (specifically, the variational "exponential" expansion used in practical calculations) is discussed in detail in Sections 1 and 2. The inverse iteration method, which is considered to be one of the most efficient computational approaches to a discretized problem with a finite basis, is reviewed in Section 3. In the last section, the convergence of numerical calculations is investigated, and the final theoretical results for 11 states of a helium atom are given.

VARIATIONAL METHOD

Let us first formulate the variational principle for bound states and describe the variational method that defines the form of basis functions of the solution expansion and the choice of variational parameters in the construction of wave functions. This method is hereinafter referred to as the "exponential" expansion.

The Hylleraas–Undheim variational principle, which is better known in mathematics as the Ray-

leigh–Ritz variational principle, is the starting point in solving the stationary Schrödinger equation

$$H\psi = E\psi \tag{1.1}$$

for a certain Hamiltonian using variational methods. This principle is considered a versatile method for deriving an approximate solution. The problems of determining the extrema or stationary values of functionals are the basic problems of variational calculation. The essence of this method consists of substituting the problem of finding the stationary values of functionals with a fundamentally less complex problem of finding the stationary values of functions of several variables [4].

Let there be a self-adjoint operator defined within the Hilbert space for which the following boundedness condition is satisfied:

$$H \geq cI, \tag{1.2}$$

where c is a certain constant. Let us then define a functional

$$\Phi(\psi) = \frac{(\psi, H\psi)}{(\psi, \psi)}, \tag{1.3}$$

that is bounded from below by c.

Theorem 1 [4]. Let H be a self-adjoint operator that satisfies condition (1.2). Let us define

$$\mu_n(H) = \max_{\dim\chi = n-1} \min_{\substack{\Psi \in \mathcal{D}(H) \\ \Psi \in \chi^\perp}} \Phi(\Psi), \tag{1.4}$$

where $\chi^\perp$ is a subspace orthogonal to χ and $D(H)$ is the domain of operator H. One of the following assertions is then true for any fixed n:

(i) n eigenvalues (degenerate eigenvalues are counted according to their multiplicity) lying below the essential spectrum boundary are present and $\mu_n(H)$ is the n-th eigenvalue (with account of multiplicity); or

(ii) $\mu_n(H)$ is the lower boundary of the essential spectrum.

The determination of eigenvalues (i.e., the energy of bound states of the stationary Schrödinger equation) comes down to calculating the saddle points of functional (1.4). The assertion of the theorem is known as the minimax principle.

Let us now consider a method that uses the Rayleigh–Ritz variational principle to solve practical eigenvalue problems and is called the Ritz process. Let ϕ_k be a complete sequence of vectors in the Hilbert space subject to the following conditions:

(i) vectors ϕ_k belong to the domain of operator H;

(ii) vectors $\phi_1, \phi_2, \ldots, \phi_n$ are linearly independent at any n.

Let us assume that $u_n = \sum_{k=1}^n x_k\phi_k$, where x_k are scalar coefficients. Inserting u_n (at fixed n) into func-

tional $\Phi(\cdot)$, we obtain a function that depends on a finite set of parameters $\{x_n\}_1^n$:

$$\Phi(x) = \left(\sum_{i,j=1}^n a_{ij}x_ix_j\right)\bigg/\left(\sum_{i,j=1}^n b_{ij}x_ix_j\right),$$

where

$$a_{ij} = (\phi_i, H\phi_j), \quad b_{ij} = (\phi_i, \phi_j).$$

The determination of minimax solutions is thus reduced to calculating the corresponding eigenvalues of the generalized eigenvalue problem:

$$Ax = \lambda Bx \tag{1.5}$$

where matrices A and B are composed of coefficients a_{ij} and b_{ij}, respectively.

Vectors ϕ_k may depend on nonlinear parameters ω. If this is the case, problem (1.5) is solved for each fixed ω and each eigenvalue number k, $\lambda_k(\omega)$ is chosen, and this value is then minimized by all values of nonlinear parameters:

$$\lambda_k = \inf_\omega \lambda_k(\omega).$$

One important condition is satisfied for Ritz estimates:

$$\mu_k(H) \leq \lambda_k. \tag{1.6}$$

It follows from there that Ritz estimates are upper bound ones. Inequality (1.6) for basis functions dependent on nonlinear parameters follows from

$$\mu_k(H) \leq \inf_\omega \lambda_k(\omega) = \lambda_k.$$

A rigorous proof of the applicability of Theorem 1 to the problems of nonrelativistic quantum mechanics with a Hamiltonian of the form

$$H = -\sum_{i=1}^n \frac{\Delta_i}{2m_i} + V(r_1, \ldots, r_n), \tag{1.7}$$

and a potential of a sufficiently general form that includes, among others, the Coulomb potential of interparticle interaction was derived by Kato [5].

GENERALIZED HYLLERAAS EXPANSION

We use the generalized Hylleraas expansion [8] for the states with arbitrary values of total orbital moment L of the system:

$$\psi(r_1, r_2) = \sum_{l_1+l_2=\mathscr{L}} \mathcal{Y}_{LM}^{l_1l_2}(r_1, r_2) \times \left[e^{-\alpha r_1 - \beta_2 - \gamma r_{12}} \sum_{l,m,n\geq 0} C_{lmn} r_1^l r_2^m r_{12}^n\right], \tag{1.8}$$

where $\mathscr{L} = L$ for the states of "normal" spatial parity $\Pi = (-1)^L$ and $\mathscr{L} = L + 1$ for the states of "anomalous" spatial parity $\Pi = (-1)^{L+1}$. The $\mathscr{Y}_{LM}^{l_1 l_2}$ functions are regular bipolar spherical harmonics [6] that depend on two angular coordinates:

$$\mathscr{Y}_{LM}^{l_1 l_2}(r_1 r_2) = r_1^{l_1} r_2^{l_2} \{ Y_{l_1}(\hat{r}_1) \otimes Y_{l_2}(\hat{r}_2) \}_{LM},$$

and spatial parity operator $P\psi = \pi\psi$ acts on the spatial coordinates in the following way: $P(r_1, r_2) \to (-r_1, -r_2)$.

The ease of use of the $\mathscr{Y}_{LM}^{l_1 l_2}$ functions stems from the fact that they correctly reproduce the behavior of the wave function at $r_1 \to 0$ (or $r_2 \to 0$) and retain the reasonable requirement of boundedness of the function within the region of variation of variables for the expression within square brackets in Eq. (1.8).

The "normal" and "anomalous" spatial parities were designated this way for the following reasons. It can be seen from expansion (1.8) that "anomalous" parity states may decompose into clusters with angular momentum of the bound pair $l \geq 1$. In atomic physics, the ground state of a pair of particles has zero angular momentum, while the boundary of the continuous spectrum in a system of three particles is defined by the ground-state energy, the energy of the pair with the lowest energy level, or zero energy (if no bound pairs are present). It follows that bound "anomalous" parity states are located below the threshold of the cluster with the excited pair state and normally lie within the continuous spectrum of a three-particle system. Therefore, after the inclusion of any interaction operator that violates spatial parity into the Hamiltonian, these states blend into the continuous spectrum and form resonances.

The calculation of matrix elements comes down to evaluating integrals of the following form:

$$\Gamma_{lmn}(\alpha, \beta, \gamma) = \iint r_1^l r_2^m r_{12}^n e^{-\alpha r_1 - \beta r_2 - \gamma r_{12}} dr_1 dr_2 dr_{12}. \quad (1.9)$$

Differentiating with respect to α under the integral sign, we obtain the following:

$$\left(-\frac{\partial}{\partial \alpha} \right) \Gamma_{l-1, mn}(\alpha, \beta, \gamma) = \Gamma_{lmn}(\alpha, \beta, \gamma).$$

Thus, all integrals may be evaluated from Γ_{000} by simple differentiation:

$$\Gamma_{lmn}(\alpha, \beta, \gamma) = \left(-\frac{\partial}{\partial \alpha} \right)^l \left(-\frac{\partial}{\partial \beta} \right)^m \left(-\frac{\partial}{\partial \gamma} \right)^n$$

$$\times \Gamma_{000}(\alpha, \beta, \gamma) = \left(-\frac{\partial}{\partial \alpha} \right)^l \left(-\frac{\partial}{\partial \beta} \right)^m \left(-\frac{\partial}{\partial \gamma} \right)^n \quad (1.10)$$

$$\times \left[\frac{2}{(\alpha + \beta)(\beta + \gamma)(\gamma + \alpha)} \right].$$

Following [7], we then use recurrence relation

$$\Gamma_{lm}(\alpha, \beta) = \frac{1}{\alpha + \beta} \Bigg[(l\Gamma_{l-1, m} + m\Gamma_{l, m-1}) + \left(-\frac{\partial}{\partial \alpha} \right)^l \left(-\frac{\partial}{\partial \beta} \right)^m f(\alpha, \beta) \Bigg].$$

Applying it successively to each pair of variables α, β, and γ, we arrive at the recurrence scheme for integral evaluation for nonnegative values of parameters (l, m, n):

$$\Gamma_{lmn} = \frac{1}{\alpha + \beta} [l\Gamma_{l-1, m, n} + m\Gamma_{l, m-1, n} + B_{lmn}],$$

$$B_{lmn} = \frac{1}{\alpha + \beta} [l B_{l-1, m, n} + m B_{l, m-1, n} + A_{lmn}], \quad (1.11)$$

$$A_{lmn} = \delta_{l0} \frac{2(m + n)!}{(\beta + \gamma)^{m+n+1}}.$$

The fact that the A_{lmn}, B_{lmn}, and Γ_{lmn} values in relations (1.11) are positive is an important feature of these relations that makes recurrence scheme (1.11) for integral evaluation resistant to the rounding errors in computer calculations.

The averaging over angular variables for the states with a nonzero total orbital moment of the system was analyzed by Drake [8]. This averaging reduces the calculation of matrix elements to integrals (1.9). A compact and efficient recurrence scheme that implements this reduction was proposed later by Efros [9].

The efficiency of the above-described variational expansions is the highest when they are applied to systems composed of two electrons and a heavy nucleus. Let us now study this version ("exponential" expansion) in more detail. This expansion assumes the following form for S states:

$$\psi(r_1, r_2, r_{12}) = \sum_n C_n e^{-\alpha_n r_1 - \beta_n r_2 - \gamma_n r_{12}}, \quad (1.12)$$

where the parameters in the exponent are chosen in one way or another. In early studies [10] that used expansion (1.12), the obtained representation was associated with the discretization of the integral representation of the wave function

$$\psi(x_1, \ldots, x_A) = \int \varphi(x_1, \ldots, x_A; \alpha) f(\alpha) d\alpha, \quad (1.13)$$

that was proposed by Griffin and Wheeler [11] in 1957. The α_n, β_n, and γ_n parameters were chosen in accordance with various quadrature integration formulas (1.13). The systematic study of expansion (1.12) with parameters generated using pseudorandom numbers was carried out in [12]. In the proposed approach,

nonlinear parameters from Eq. (1.12) are generated using the following simple formulas:

$$\alpha_n = \left[\left\lfloor \frac{1}{2}n(n+1)\sqrt{p_\alpha} \right\rfloor (A_2 - A_1) + A_1\right],$$

$$\beta_n = \left[\left\lfloor \frac{1}{2}n(n+1)\sqrt{p_\beta} \right\rfloor (B_2 - B_1) + B_1\right], \qquad (1.14)$$

$$\gamma_n = \left[\left\lfloor \frac{1}{2}n(n+1)\sqrt{p_\gamma} \right\rfloor (C_2 - C_1) + C_1\right],$$

where $\lfloor x \rfloor$ is the fractional part of x and p_α, p_β, and p_γ are certain prime numbers. These simple generators of pseudorandom numbers have their advantage in the reproducibility of the results of variational calculations. The convergence rate of the exponential expansion with a pseudorandom strategy for choosing non-linear parameters (1.14) is exceptionally high at the sets of basis functions of moderate dimensionalities (up to 100−200 test functions). Rapid basis degeneration that results in the loss of computational stability in the double precision arithmetic by basis dimensionality $N = 200$ is among the disadvantages of the method.

Let us write out for convenience the exponential variational expansion in its complete form with account for the angular dependence of the wave function that describes the rotational degrees of freedom:

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \sum_{l_1 + l_2 = \mathcal{L}} \mathcal{Y}_{LM}^{l_1 l_2}(\hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2) G_{l_1 l_2}^{L\pi}(r_1, r_2, r_{12}),$$

$$G_{l_1 l_2}^{L\pi}(r_1, r_2, r_{12}) = \sum_n C_n e^{-\alpha_n r_1 - \beta_n r_2 - \gamma_n r_{12}}, \qquad (1.15)$$

where $\mathcal{L} = L$ or $L + 1$ (depending on the spatial parity of the state) and the complex parameters in the exponent are generated in a pseudorandom way (1.14).

It was already noted that the convergence rate is reduced when the ground state of a helium atom is calculated. This may be attributed to the fact that the wave function has a logarithmic singularity at $r_1, r_2 \to 0$: $\rho^2 \ln\rho$, where $\rho = \sqrt{r_1^2 + r_2^2}$ is the hyperradius of two electrons [13]. In order to remedy the situation, one should construct a multilayer variational expansion composed of several independent sets of basis functions, the optimum variational nonlinear parameters for which are found independently. Thus, each set of basis functions defines the optimum approximation in a certain region of coordinates of the system. In the case of a helium atom, the regions should be enclosed within each other and be more and more compact in terms of the hyperradius ($\rho < \rho_n = a^n$, where $a \approx 0.1$ and $n = 1, 2, 3, \ldots$). This strategy makes the exponential expansion an efficient and versatile solution method for bound states in the quantum three-body problem with Coulomb interaction. The capabilities of this method were demonstrated in [14, 15].

INVERSE ITERATION METHOD

It was shown in Section 1 that the stationary Schrödinger equation is reduced to the generalized symmetrical eigenvalue problem with the help of the Ritz procedure:

$$Ax = \lambda Bx, \qquad (1.16)$$

where A is a symmetric matrix and B is a symmetric positive-definite matrix. The standard diagonalization procedure may be used to solve Eq. (1.16). In order to do that, matrix $B = L \cdot L^T$ is expanded into a product of upper and lower triangular matrices, and the problem is reduced to the standard symmetrical eigenvalue problem:

$$A'y = \lambda y, \qquad (1.17)$$

and

$$A'y = L^{-1}AL^{-T}, \quad x = L^T y. \qquad (1.18)$$

However, this method is too laborious ($\sim 20N^3$ multiplication operations) and is less resistant to calculation errors. If only a single eigenvalue (eigenvector) is needed, the solution may be obtained efficiently ($\sim N^3/6$ multiplication operations) with the help of the inverse iteration method:

$$(A - \mu)x_k^{(n+1)} = s^{(n)}x_k^{(n)}, \qquad (1.19)$$

where scalar factor $s^{(n)}$ is chosen in such a way that $\left\| x_k^{(n+1)} \right\| = 1$. If μ is close to exact eigenvalue λ_k, vector sequence $x_k^{(n)}$ converges rapidly to exact eigenvector x_k, and $\lambda_k^{(n)} = (x_k^{(n)}, Ax_k^{(n)})$ converges rapidly to exact value λ_k.

In order to illustrate this, one may assume, without a loss of generality, that matrix A is a diagonal one. The solution may then be written down in the explicit form:

$$x_k^{(n)} = c_n \left(\left(\frac{\lambda_k - \mu}{\lambda_1 - \mu} \right)^n u_1, \ldots, u_k, \ldots, \left(\frac{\lambda_k - \mu}{\lambda_n - \mu} \right)^n u_n \right)^T. \quad (1.20)$$

It can be seen from Eqs. (1.20) that all components of vector $x_k^{(n)}$ (except for u_k, which remains equal to unity) tend to zero under the given normalization conditions. Practical calculations demonstrate that this method is also the most resistant to rounding errors (calculation errors).

RESULTS AND DISCUSSION

The results of numerical calculations of the ionization energies for S, P, and D states of a helium atom are listed in Table 1. These calculations were carried out using the inverse iteration method. Variational parameters were optimized manually. It should be noted that the optimum variational parameters for dif-

Table 1. Nonrelativistic energy levels of S, P, and D states of a helium atom. N is the number of basis functions

State	Basis (N)	E_{nr}
1^1S	2000	−2.903 724 377 034 119 598 28
1^1S	3000	−2.903 724 377 034 119 598 31
2^1S	2000	−2.145 974 046 054 417 415 77
2^1S	3000	−2.145 974 046 054 417 415 81
2^3S	1500	−2.175 229 378 236 791 305 74
2^3S	2500	−2.175 229 378 236 791 305 74
2^1P	3000	−2.123 843 086 498 101 359 24
2^1P	4000	−2.123 843 086 498 101 359 25
2^3P	3000	−2.133 164 190 779 283 205 11
2^3P	4000	−2.133 164 190 779 283 205 14
3^1S	2500	−2.061 271 989 740 908 650 15
3^1S	3000	−2.061 271 989 740 908 650 72
3^3S	2500	−2.068 689 067 472 457 192 00
3^3S	3000	−2.068 689 067 472 457 192 00
3^1P	3000	−2.055 146 362 091 943 536 74
3^1P	4000	−2.055 146 362 091 943 536 89
3^3P	2500	−2.058 081 084 274 275 331 26
3^3P	3500	−2.058 081 084 274 275 331 34
3^1D	3000	−2.055 620 732 852 246 489 39
3^1D	4000	−2.055 620 732 852 246 489 39
3^3D	3000	−2.055 636 309 453 261 327 11
3^3D	4000	−2.055 636 309 453 261 327 11

Table 2. Investigation of the convergence of the nonrelativistic energy of 1^1S states of a helium atom

Basis (N)	E_{nr}	ΔE_{nr}
1600	−2.903 724 377 034 119 597 961	
1800	−2.903 724 377 034 119 598 225	264
2000	−2.903 724 377 034 119 598 282	57
2400	−2.903 724 377 034 119 598 307	25
3000	−2.903 724 377 034 119 598 311	4

Table 3. Investigation of the convergence of the nonrelativistic energy of 2^1P states of a helium atom

Basis (N)	E_{nr}	ΔE_{nr}
2000	−2.123 843 086 498 101 358 687	
2500	−2.123 843 086 498 101 359 180	493
3000	−2.123 843 086 498 101 359 237	57
4000	−2.123 843 086 498 101 359 246	9

CONCLUSIONS

Variational wave functions of bound states were obtained by solving the Schrödinger equation for the quantum three-body problem with Coulomb interaction using a variational approach based on exponential expansion with the parameters of exponents being chosen in a pseudorandom way. The results of calculations of the nonrelativistic energy levels for a helium atom were presented. The numerical calculation results are listed in Table 1. The convergence was studied as a function of the number of test functions. The results of these studies demonstrated that the energy values were accurate to 19−20 significant digits. This accuracy allows one to obtain reliable theoretical predictions.

ferent states differ from each other, and the calculation accuracy depends to a considerable extent (5−8 digits) on the choice of the optimum variational parameters for the given bound state. Bases with N = 1500, 2000, 2500, and 3000 functions were used to optimize the variational parameters. When the states listed in the table were calculated, 3−5 "layers" of basis functions were used.

Program modules of quadruple and sextuple precision (32 and 48 decimal digits, respectively) that were developed by one of the authors of the present paper were used in order to remedy the problem of the numerical instability of calculations at large values of N.

The convergence of the nonrelativistic energy value is studied in Tables 2 and 3 as a function of number N of basis functions. The difference between two neighboring values (calculated for the last digits given in Tables 2 and 3) is listed in the third column. It is seen clearly that the convergence rate is high. This allows one to obtain highly accurate results.

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