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Two-Centre Corrections to the Spherical and Parabolic Bases of the Hydrogen Atom

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Abstract

A brief analysis of the fundamental bases of the hydrogen atom, which are the eigenfunctions of its Hamiltonian and of the one of the generators of the hidden symmetry group SO (4) is carried out. The spheroidal corrections to the spherical and parabolic bases up to the corrections of third order are calculated by means of perturbation theory. These expansions were analysed in terms of additional integrals of motion.

1 Introduction

The study of systems with hidden symmetry in spaces of constant curvature has a rich history. Starting from the works of Laplace [1], Runge [2] and Lenz [3], in which the additional integral of motion was firstly proposed, namely Laplace-Runge-Lenz vector, the theory of hidden symmetries has found a large variety of applications. The first succesfull applying of this theory to the quantum-mechanical problems was carried out by Pauli [4] for the calculation of the hydrogen atom discrete energy spectrum [5]. It is well known, that the quantum-mechanical quantization problem for hydrogen atom or hydrogen-like ion allows the separation of variables in spherical, parabolic and prolate spheroidal coordinate systems. Since the energy levels in this problem for every coordinate system are the same, only the wave functions must be considered.

A wide variety of problems makes it necessary to work with different coordinate systems and use the interbasis expansions. For instance, a spherical coordinate system is commonly used for the spectroscopy of hydrogen-like systems, while for the study of the Stark effect the parabolic coordinate system is more preferable. On the other hand, the spheroidal coordinate system is the most convenient for the study of the electron motion in a field of two fixed Coulomb centres. Coulomb spheroidal wave functions obtained herewith were determined by many authors both directly, using explicit form of basic functions [6, 7, 8], and obliquely, i.e.

through the additional motion integrals [9, 10]. For fixed n and m (here n is the principal quantum number, m is the magnetic quantum number) the problem reduces to finding the solution of the linear homogeneous algebraic equation system of order $n - |m|$. Moreover, for small $n - |m|$ this system of equations can be solved analytically. However, for arbitrary $n - |m|$ and R (here R is the distance between the foci of the spheroidal coordinate system) the general solution is still unavailable.

Coulomb spheroidal wave functions can be determined in general form in two limiting cases of large and small internuclear distances R. In the first case, these functions can be represented in the form of linear combination of Coulomb parabolic wave functions [6, 11] when in the second case – in the form of linear combination of Coulomb spherical wave functions [11].

In this paper, the spheroidal corrections to the spherical and parabolic bases of hydrogen atom for small and large R were obtained. Our approach is based on the employing the standard scheme of the Rayleigh-Schrödinger perturbation theory and on the taking into account the additional integrals of motion. In Sec. 2, we consider the additional motion integrals and the spherical, parabolic and spheroidal bases of hydrogen atom. In Sec. 3, the spheroidal corrections to the spherical and parabolic bases of hydrogen atom were calculated in the framework of the perturbation theory. We also show that in each order of our approach the corrections to the Coulomb spheroidal wave functions are expressed in a finite number of the basic functions of the corresponding representation.

2 Spherical, parabolic and spheroidal bases of hydrogen atom

The one-centre Coulomb problem allows to separate the variables in three coordinate systems: spherical, parabolic and prolate spheroidal, where the first two of them are the limiting cases of the third one (see §1 of [6]). Thus, the solutions of hydrogen-like atom problem in spheroidal coordinates can be found without considering the equations for Coulomb spheroidal quasiradial and quasiangular wave functions but using the interbasis expansions. Here we will construct the solutions of the hydrogen-like atom problem using the additional motion integrals (constant of motion) and known solutions of this problem in other coordinate systems [10, 12]. For this purpose let us first consider the spherical Ψ^{sph}_{nlm}, parabolic $\Psi^{par}_{n_1n_2m}$ and spheroidal Ψ^{spher}_{nqm} bases of hydrogen atom for discrete spectrum.

A. Spherical basis. The hydrogen atom wave function Ψ^{sph}_{nlm} in spherical coordinate system is an eigenfunction of the following operators (hereinafter we use the atomic system of units: $e = m_e = \hbar = 1$):

$$\hat{H}\Psi^{sph}_{nlm} = E_n\Psi^{sph}_{nlm}, \quad \hat{\vec{L}}^2\Psi^{sph}_{nlm} = l(l+1)\Psi^{sph}_{nlm}, \quad \hat{L}_z\Psi^{sph}_{nlm} = \pm m\Psi^{sph}_{nlm}. \quad (1)$$

Here $\hat{\vec{L}} = [\vec{r}\times\hat{\vec{p}}]$ and $\hat{L}_z$ are the orbital angular momentum operator and operator of its projection to the intercentre axis $\vec{R}$ (centres are the foci of the prolate

spheroidal coordinate system), respectively, and $\hat{H}$ is the Hamiltonian of the one-centre Coulomb problem:

$$\hat{H} = \frac{\hat{\vec{p}}^2}{2m} - \frac{1}{r}, \qquad E_n = -\frac{1}{2n^2}, \qquad n = l + n_r + 1. \tag{2}$$

In (1), (2) the numbers $n > l \geq m$ are integers, l is the azimuthal quantum number.

B. Parabolic basis. The hydrogen atom wave function $\Psi^{par}_{n_1 n_2 m}$ in parabolic coordinate system is an eigenfunction of the operators

$$\hat{H}\Psi^{par}_{n_1 n_2 m} = E_n \Psi^{par}_{n_1 n_2 m}, \;\; \hat{A}_z \Psi^{par}_{n_1 n_2 m} = (n_1 - n_2)\Psi^{par}_{n_1 n_2 m}, \;\; \hat{L}_z \Psi^{par}_{n_1 n_2 m} = \pm m \Psi^{par}_{n_1 n_2 m}, \tag{3}$$

where non-negative integers n_1, n_2 are the parabolic quantum numbers so that $n = n_1 + n_2 + m + 1$. The appearing in (3) operator is the Laplace-Runge-Lenz vector

$$\hat{\vec{A}} = n\left\{\frac{1}{2}[\hat{\vec{L}} \times \hat{\vec{p}}] - \frac{1}{2}[\hat{\vec{p}} \times \hat{\vec{L}}]\frac{\vec{r}}{r}\right\}, \tag{4}$$

which commutes with the Hamiltonian $\hat{H}$ of the hydrogen atom and thus it is the integral of motion.

C. Spheroidal basis. The hydrogen atom wave function Ψ^{spher}_{nqm} in prolate spheroidal coordinate system is an eigenfunction of the operators

$$\hat{H}\psi^{spher}_{nqm} = E_n \psi^{spher}_{nqm}, \quad \hat{\Lambda}\psi^{spher}_{nqm} = \lambda_q \psi^{spher}_{nqm}, \quad \hat{L}_z \psi^{spher}_{nqm} = \pm m \psi^{spher}_{nqm}, \tag{5}$$

where k and q – spheroidal quantum numbers, and $k + q + m + 1 = n$. The separation of variables in the prolate spheroidal system [6] for the considered Coulomb problem is possible because along with the hamiltonian $\hat{H}$ and the projection of the orbital angular momentum operator $\hat{L}_z$ there is an additional integral of motion here – the separation constant, operator of which has the following form:

$$\hat{\Lambda} = -\hat{\vec{L}}^2 - \frac{R}{n}\hat{A}_z. \tag{6}$$

Here R is a parameter of the spheroidal system (the distance between the foci).

From the properties of the SO(4) algebra it follows that nonzero matrix elements of the third component $\hat{A}_z$ of the Laplace-Runge-Lenz vector $\hat{\vec{A}}$ (4) in spherical basis are determined by the following formula [13]:

$$\left(\hat{A}_z\right)_{l'l} = \int \psi^{\tilde{n}\hat{0}*}_{nl'm} \hat{A}_z \psi^{\tilde{n}\hat{0}}_{nlm} dV = -\left\{\frac{(l+|m|)(l-|m|)(n-l)(n+l)}{(2l+1)(2l-1)}\right\}^{1/2} \delta_{l',l-1}$$
$$-\left\{\frac{(l+|m|+1)(l-|m|+1)(n-l-1)(n+l+1)}{(2l+1)(2l+3)}\right\}^{1/2} \delta_{l',l+1}. \tag{7}$$

Let us write the transformations connecting the spherical and parabolic wave functions of hydrogen atom for a discrete spectrum [12, 13]:

$$\psi^{par}_{n_1n_2m} = (-1)^{n_2+\frac{m+1}{2}} \sum_{l=|m|}^{n-1} (-1)^l C^{l,|m|}_{\frac{n-1}{2},\frac{n_1-n_2+|m|}{2};\frac{n-1}{2},\frac{n_2-n_1+|m|}{2}} \psi^{sph}_{nlm}, \quad (8)$$

$$\psi^{sph}_{nlm} = (-1)^{l+\frac{m-|m|}{2}} \sum_{n_2=0}^{n-|m|-1} (-1)^{n_2} C^{l,|m|}_{\frac{n-1}{2},\frac{n-1}{2}-n_2;\frac{n-1}{2},n_2+|m|-\frac{n-1}{2}} \psi^{par}_{n_1n_2m}, \quad (9)$$

where $C^{l,|m|}$ are the Clebsch-Gordan coefficients. These transformations reduce the calculation of matrix elements of operator $\hat{\vec{L}}^2$ on parabolic functions $\psi^{par}_{n_1n_2m}$ (3) to the simpler problem of finding the matrix elements of the same operator $\hat{\vec{L}}^2$ but on spherical basis (1). The resulting expression for $\left(\hat{\vec{L}}^2\right)_{n_2\,n_2'}$ is of the form [12]

$$\begin{aligned}\left(\hat{\vec{L}}^2\right)_{n_2\,n_2'} &= [(n_2+1)(n-|m|-n_2-1)+(n-n_2)(n_2+|m|)]\,\delta_{n_2',n_2} \\ &- [(n_2+1)(n-|m|-n_2-1)(n-n_2-1)(n_2+|m|+1)]^{1/2}\,\delta_{n_2',n_2+1} \\ &- [n_2(n-|m|-n_2)(n-n_2)(n_2+|m|)]^{1/2}\,\delta_{n_2',n_2-1}. \end{aligned} \quad (10)$$

Finally, we give the transformation from the spheroidal basis (5) to a parabolic one (3):

$$\psi^{spher}_{nqm} = \sum_{n_2=0}^{n-|m|-1} U^{n_2}_{nqm} \psi^{par}_{n_1n_2m}. \quad (11)$$

In [12], the following three-term recurrence relation for the coefficients $U^{n_2}_{nqm}$ was obtained:

$$\begin{aligned}&\left[\lambda_q + (n_2+1)(n-|m|-n_2-1)+(n-n_2)(n_2+|m|)\right. \\ &\left.+\frac{R}{n}(n-|m|-2n_2-1)\right] U^{n_2}_{nqm} = [n_2(n-|m|-n_2)(n-n_2)(n_2+|m|)]^{1/2}\,U^{n_2-1}_{nqm} \\ &+[(n_2+1)(n-|m|-n_2-1)(n-n_2-1)(n_2+|m|+1)]^{1/2}\,U^{n_2+1}_{nqm}. \end{aligned} \quad (12)$$

The integrals of motion considered above along with the Hamiltonian $\hat{H}(2)$ of the hydrogen atom form the quadratic algebra. In the case of fixed negative energy values, the motion integrals form an algebra SO(4), and in the case of positive values of energy – algebra SO(3.1). Due to the hidden symmetry, the hydrogen atom problem allows the separation of variables not only in spherical and parabolic coordinate systems, but also in spheroidal one.

3 The Rayleigh-Schrödinger perturbation theory

Consider the Sturm-Liouville equation for operator $\hat{\Lambda}$ in the case of small R:

$$\left(-\hat{\vec{L}}^2 - \frac{R}{n}\hat{A}_z\right)\psi_{nqm}^{spher} = \lambda_q \psi_{nqm}^{spher}. \tag{13}$$

When $R \ll 1$, the second term in the left part of (13) can be considered as a small perturbation and the spheroidal basis forms the unperturbed zeroth-order functions. Considering the explicit form (7) of matrix elements of operator $\hat{A}_z$ and the formulae of the standard Rayleigh-Schrödinger perturbation theory [5], we find the following expressions for the eigenvalues $\lambda_q(R)$ and eigenfunctions ψ_{nqm}^{spher} including the terms of third order:

$$\lambda_q(R) = -l(l+1) + \frac{R^2}{2n^2}\left[\frac{A_{n,l+1}B_{l+1,m}}{(l+1)} - \frac{A_{n,l}B_{l,m}}{l}\right] + \mathrm{O}(R^4), \tag{14}$$

$$\begin{aligned}
\psi_{nqm}^{spher} &= \psi_{nlm}^{sph} - \frac{R}{2n}\frac{\sqrt{A_{n,l}B_{l,m}}}{l}\psi_{n,l-1,m}^{sph} + \frac{R}{2n}\frac{\sqrt{A_{n,l+1}B_{l+1,m}}}{(l+1)}\psi_{n,l+1,m}^{sph} \\
&- \frac{R^2}{8n^2}\left[\frac{A_{n,l+1}B_{l+1,m}}{(l+1)^2} + \frac{A_{n,l}B_{l,m}}{l^2}\right]\psi_{nlm}^{sph} + \frac{R^2}{4n^2}\frac{\sqrt{A_{n,l-1}A_{n,l}B_{l-1,m}B_{l,m}}}{l(2l-1)}\psi_{n,l-2,m}^{sph} \\
&+ \frac{R^2}{4n^2}\frac{\sqrt{A_{n,l+2}A_{n,l+1}B_{l+2,m}B_{l+1,m}}}{(2l+3)(l+1)}\psi_{n,l+2,m}^{sph} \\
&- \frac{R^3}{24n^3 l(l-1)}\sqrt{\frac{A_{n-2,l}A_{n-1,l}A_{n,l}B_{l-2,m}B_{l-1,m}B_{l,m}}{(2l+1)(2l-3)(2l-1)^3}}\psi_{n,l-3,m}^{sph} \\
&+ \frac{R^3}{24n^3}\frac{\sqrt{A_{n,l+3}A_{n,l+2}A_{n,l+1}B_{l+3,m}B_{l+2,m}B_{l+1,m}}}{(l+1)(l+2)(2l+3)}\psi_{n,l+3,m}^{sph} \\
&+ \frac{R^3\sqrt{A_{n,l+1}B_{l+1,m}}}{16n^3(l+1)^3}\left[\frac{2(l+1)A_{n-1,l}B_{l+2,m}}{(2l+3)(2l+5)} - \frac{l^2-1}{l^2}A_{n,l}B_{l,m}\right. \\
&\left. + 3A_{n,l+1}B_{l+1,m}\right]\psi_{n,l+1,m}^{sph} - \frac{R^3\sqrt{A_{n,l}B_{l,m}}}{16n^3 l^3}\left[\frac{2l^2 A_{n,l-1}B_{l-1,m}}{2l-1} - 3A_{n,l}B_{l,m}\right. \\
&\left. + \frac{l(l+2)A_{n,l+1}B_{l+1,m}}{(l+1)^2}\right]\psi_{n,l-1,m}^{sph} + \mathrm{O}(R^4),
\end{aligned} \tag{15}$$

where

$$A_{n,l} = \frac{n^2 - l^{\,2}}{2l+1}, \qquad B_{l,m} = \frac{l^2 - m^2}{2l-1}.$$

Note that the corrections to the eigenvalues of the first $\lambda_q^{(1)} \sim R^1$ and third $\lambda_q^{(3)} \sim R^3$ orders are equal to zero due to the fact that the matrix element $(A_z)_{l\,l}$ for corrections of all odd powers is also equal to zero.

Consider now the case of large values of R. Multiplying (13) by n/R we arrive to the following equation

$$\left(\hat{A}_z - \frac{n}{R}\hat{\vec{L}}^2\right)\psi_{nqm}^{spher} = \frac{n}{R}\lambda_q\psi_{nqm}^{spher}. \tag{16}$$

The role of the perturbation here plays the second term $n\hat{\vec{L}}^2/R$ in the left part of (16). Thus, the parabolic basis acquires the status of undisturbed wave functions in the framework of the perturbation theory and all higher-order corrections then can be represented by the matrix elements on the parabolic wave functions (see (11)). Considering the expressions (10) for the matrix elements of the perturbation operator $n\hat{\vec{L}}^2/R$ and applying the standard scheme of the Rayleigh-Schrödinger perturbation theory [5] up to the terms of third order we obtain the following results for the eigenvalues $\lambda_q(R)$ and the wave functions ψ_{nqm}^{spher} in the form of series in R^{-1}:

$$\begin{aligned}
\lambda_q(R) &= -\frac{R}{n}\,(n-|m|-2n_2-1) - (n_2+1)(n-|m|-n_2-1)\\
&- (n-n_2)(n_2+|m|) + \frac{n}{2R}\Big[n_2(n-|m|-n_2)(n-n_2)(n_2+|m|)\\
&- (n_2+1)(n-|m|-n_2-1)(n-n_2-1)(n_2+|m|+1)\Big]\\
&- \frac{n^2}{2R^2}\Big[(2n_2-n-|m|)n_2(n-|m|-n_2)(n-n_2)(n_2+|m|)\\
&- (2n_2-n+|m|+2)(n_2+1)(n-|m|-n_2-1)(n-n_2-1)(n_2+|m|+1)\Big],
\end{aligned} \tag{17}$$

$$\begin{aligned}
\psi_{nqm}^{spher} &= \psi_{n_1n_2m}^{par} - \frac{n}{2R}\sqrt{C_{n_2+\frac{1}{2},|m|}}\,\psi_{n_1-1,n_2+1,m}^{par} + \frac{n}{2R}\sqrt{C_{n_2-\frac{1}{2},|m|}}\,\psi_{n_1+1,n_2-1,m}^{par}\\
&- \frac{n^2}{8R^2}\left[C_{n_2+\frac{1}{2},|m|} + C_{n_2-\frac{1}{2},|m|}\right]\psi_{n_1n_2m}^{par}\\
&+ \frac{n^2}{4R^2}\Big\{\sqrt{C_{n_2-\frac{1}{2},|m|}}\,[(n-n_2)(2n-2n_2-|m|-3)+2(n_2+|m|)]\,\psi_{n_1+1,n_2-1,m}^{par}\\
&+ \sqrt{C_{n_2+\frac{1}{2},|m|}}\,[(n-n_2)(2n-2n_2-|m|-3)+2(n_2+|m|+2)]\,\psi_{n_1-1,n_2+1,m}^{par}\Big\}\\
&+ \frac{n^2}{8R^2}\left[\sqrt{C_{n_2+\frac{1}{2},|m|}C_{n_2+\frac{3}{2},|m|}}\,\psi_{n_1-2,n_2+2,m}^{par} + \sqrt{C_{n_2-\frac{1}{2},|m|}C_{n_2-\frac{3}{2},|m|}}\,\psi_{n_1+2,n_2-2,m}^{par}\right]\\
&- \frac{n^3}{4R^3}\Big[C_{n_2+\frac{1}{2},|m|}(2n_2-n+|m|+2) + C_{n_2-\frac{1}{2},|m|}(2n_2-n+|m|)\Big]\psi_{n_1n_2m}^{par}\\
&+ \frac{n^2}{16R^3}\sqrt{C_{n_2+\frac{1}{2},|m|}}\Big[8(2n_2-n+|m|+2)^2 + C_{n_2-\frac{1}{2},|m|} - 3C_{n_2+\frac{1}{2},|m|}\\
&+ C_{n_2+\frac{3}{2},|m|}\Big]\psi_{n_1-1,n_2+1,m}^{par} - \frac{n^3}{16R^3}\sqrt{C_{n_2-\frac{1}{2},|m|}}\Big[8(2n_2-n+|m|)^2\\
&+ C_{n_2+\frac{1}{2},|m|} - 3C_{n_2-\frac{1}{2},|m|} + C_{n_2-\frac{3}{2},|m|}\Big]\psi_{n_1+1,n_2-1,m}^{par}
\end{aligned}$$

$$+\frac{n^3}{8R^3}[4n_2-2n+2m+5]\sqrt{C_{n_2+\frac{1}{2},|m|}C_{n_2+\frac{3}{2},|m|}}\psi^{par}_{n_1-2,n_2+2,m}$$
$$+\frac{n^3}{8R^3}[4n_2-2n+2m-1]\sqrt{C_{n_2-\frac{1}{2},|m|}C_{n_2-\frac{3}{2},|m|}}\psi^{par}_{n_1+2,n_2-2,m}$$
$$+\frac{n^3}{48R^3}\sqrt{C_{n_2+\frac{1}{2},|m|}C_{n_2+\frac{3}{2},|m|}C_{n_2+\frac{5}{2},|m|}}\psi^{par}_{n_1-3,n_2+3,m}$$
$$-\frac{n^3}{48R^3}\sqrt{C_{n_2-\frac{1}{2},|m|}C_{n_2-\frac{3}{2},|m|}C_{n_2-\frac{5}{2},|m|}}\psi^{par}_{n_1+3,n_2-3,m}, \qquad (18)$$

where

$$C_{n_2,|m|}=(n_2+1/2)(n-|m|-n_2-1/2)(n-n_2-1/2)(n_2+|m|+1/2).$$

The formulae (14)–(15) and (17)–(18) were obtained in the framework of third-order perturbation theory. Note also that in the formula for ψ^{spher}_{nqm} at large R presented in [12] the second and the third terms must be four times smaller.

The main results of this paper are the interbasis expansions (14)–(15) and (17)–(18). They can be used in solving various problems of atomic and molecular physics, for instance, the $1/R$-expansions (17), (18) are applicable for construction of the two-centred wave function near the remote nucleus [14], which plays the crucial role in the two-electron exchange processes.

Conclusions

A brief analysis of the hydrogen atom fundamental bases, which are the eigenfunctions of the Hamiltonian $\hat{H}(2)$, and of the one of the generators of the hidden symmetry group SO(4) was carried out. The expansions of one of the fundamental bases in the other ones were examined in terms of additional integrals of motion, allowing to calculate the spheroidal corrections to the spherical and parabolic bases of the hydrogen atom at small and large intercentric distances R. It was also shown that in every order of the perturbation theory the corrections to the wave function are expressed in terms of a finite number of basis functions.

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