Circular Rydberg States of Atomic Hydrogen in an Arbitrary Magnetic Field

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Abstract We report a theoretical scheme using a B-spline basis set to improve the poor computational accuracy of circular Rydberg states of hydrogen atoms in the intermediate magnetic field. This scheme can produce high accuracy energy levels and valid for an arbitrary magnetic field. Energy levels of hydrogen are presented for circular Rydberg states with azimuthal quantum numbers $|m| = 10–70$ as a function of magnetic field strengths ranging from zero to $2.35 \times 10^9$ T. The variation of spatial distributions of electron probability densities with magnetic field strengths is discussed and competition between Coulomb and magnetic interactions is illustrated.

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Key words: hydrogen atom, circular Rydberg state, magnetic field, B-spline basis

1 Introduction

Circular states of Rydberg atoms are states with $|m| = n - 1$, where $m$ and $n$ are the azimuthal and principal quantum numbers, respectively. The word “circular” here is used because the classical orbits corresponding to these quantum states are circular. Such states have drawn considerable interests since the early development stage of quantum theory. In modern physics, circular Rydberg states play a crucial role in understanding the correspondence limit between classical and quantum physics.

Rydberg atoms in circular states possess some special properties, such as the largest magnetic moment, the smallest Stark effect, the longest radiative lifetime, and highly anisotropic collision cross sections. These properties make them applicable in many research domains, including cavity QED, quantum computation, and information processing. Measurements of fundamental physics constants, ultrafast dynamics of Rydberg atoms, and so on. The earliest experiment for circular state populations was made by Hulet and Kleppner. In that experiment, the circular states of lithium atoms with $|m| = 17, 18$ were prepared using an adiabatic microwave transfer technique. Soon afterwards, the same technique was applied to the other two alkali metal atoms, cesium and rubidium. The external electric and magnetic fields have been adopted to control adiabatic microwave transfer and to split $\pm n$ degeneracy, respectively. A theoretical scheme using crossed electric and magnetic fields to populate circular states was proposed by Delende and Guy. The creation of circular states of hydrogen atoms was based on both adiabatic microwave transfer and the crossed fields method. Very recently, Maeda et al. successfully made long-lived, nondispersing Bohr wave packets starting from Li Rydberg atoms in circular states. Obviously interests in circular states have been enhanced in recent years.

Besides fundamental physics interests, investigation of magnetized H atoms is of application importance in astronomy and astrophysics. To understand the behavior of magnetized white dwarf stars ($10^2–10^5$ T) and neutron stars ($10^7–10^9$ T), the accurate knowledge of the physical properties of atoms in strong magnetic fields is essential.

Much attention has been paid to experiment and astronomical observation, but corresponding computations of circular Rydberg states of atoms in external fields are relatively rare. Wunner et al. solved the Schrödinger equations for circular states of magnetized hydrogen atoms. For low fields, they expanded wave functions in terms of a complete, orthonormal set of functions in the radial direction and spherical harmonics in the angular direction, while for high fields ($\leq 10^7$ T), they employed the adiabatic approximation. In the intermediate field range, they smoothly jointed the two-side results. Seemingly, the method of Wunner et al. may effectively handle any magnetic field from low to high. However, that is not true. It is widely known that the adiabatic approximation is valid only for extremely high magnetic fields, and interpolation in the intermediate field range may produce poor results because of the used energies stemming from the adiabatic approximation. Here it is necessary to point out that intermediate magnetic field used in literature is a very ambiguous concept. Strictly speaking in physics, the “intermediate magnetic field” means a magnetic field when Coulomb and magnetic interactions are of a comparable magnitude. In other words, also it is closely relevant to the energy levels besides magnetic fields.
Liu et al.\textsuperscript{[15]} performed calculations of circular Rydberg states of H atoms in magnetic field strengths ranging from zero to 70.5 T, based on a B-spline expansion method, in which wave functions are expanded in terms of B-spline functions in both the radial and angular directions. However, their method is limited to low magnetic fields. It should be emphasized, again, that the ambiguous concept of the so-called low magnetic fields is being used here. The disadvantages of their method have been analyzed recently in Ref. \[16\]. The accuracy of their energy levels decreases with magnetic fields $B$ when $B > 47$. Gernemann, et al.\textsuperscript{[17]} developed a dimensional perturbation theory to evaluate circular Rydberg states of magnetized atomic hydrogen with an extreme high accuracy. The theory is applicable to entire range of magnetic field strengths, but is limited to high azimuthal quantum number ($m \gg 1$). Later, Watson and coworker\textsuperscript{[18]} extended their method to include low-$m$ situations. However, not in all field strengths, circular states may be treated because of the limitation of this theory.

This paper focuses on the problem of circular Rydberg states in the intermediate magnetic fields. First of all, we develop a method to evaluate circular Rydberg states of atomic hydrogen in a strong magnetic field. In cylindrical coordinates, the wave functions are expanded in terms of Landau states in the $\rho$ direction and the B-spline basis in the $z$ direction. This method is combined together with a recently reported finite-basis-set technique\textsuperscript{[16]} which is valid for weak fields. We will show that the combination scheme is a high accuracy tool to compute circular Rydberg states of atomic hydrogen in the intermediate magnetic fields, and valid for an arbitrary magnetic field.

2 Theoretical Method

The nonrelativistic Hamiltonian of hydrogen atoms on a uniform magnetic field along the $z$ axis reads in atomic units

$$\hat{H} = -\frac{1}{2} \nabla^2 - \frac{1}{\sqrt{\rho^2 + z^2}} + \gamma \left( \ell_z + 2s_z \right) + \frac{\gamma^2}{8} \rho^2 ,$$

(1)

where $\gamma$ is the magnetic field strength in units of $B_0 \approx 2.35 \times 10^5$ T, $\ell_z$ and $s_z$ are the $z$ components of the orbital and spin angular momenta, respectively, the third term linear in $\gamma$ is the paramagnetic potential, and the fourth term quadratic in $\gamma$ is the diamagnetic potential. It is easily seen that $\hat{H}$ commutes with $\ell_z$ (corresponding quantum number $m$) as well as $s_z$. Depending on relative magnitudes of Coulomb and diamagnetic potentials, the symmetry of the system changes. The system is more spherically symmetric if the Coulomb potential is dominant, and more cylindrically symmetric if the diamagnetic potential is dominant.

For strong magnetic fields, we solve the Schrödinger equation in cylindrical coordinates by expanding wave functions in terms of Landau states in the $\rho$ direction and the B-spline basis in the $z$ direction. The wave function is expressed as

$$\Psi(\rho, z, \phi) = \sum_{i, n} C_{i,n} B_i(z) \mathcal{R}_n(\rho, \phi) ,$$

(2)

where $\mathcal{R}_n(\rho, \phi)$ is the normalized wave function for the Landau state with $n \geq 0$, given by\textsuperscript{[19]}

$$\mathcal{R}_n(\rho, \phi) = \mathcal{R}_n(\rho) \frac{e^{im\phi}}{\sqrt{2\pi}} \frac{\Gamma(|m|+1/2)}{\Gamma(|m|+n)!} \sqrt{|m|+n}! \left( \frac{\rho}{\gamma} \right)^{|m|+n} \exp\left[-\frac{\rho^2}{2\gamma^2} - \frac{\gamma}{\rho^2} \right] ,$$

(3)

with $1 F_1$ being the confluent hypergeometric function, and $B_i(z)$ are the B-spline functions. The B-spline functions along with their properties have been outlined in Ref. \[16\], and a complete description can be also found in the book of de Boor.\textsuperscript{[20]} Therefore, here it is unnecessary to give any elaboration once again. $\mathcal{R}_n(\rho, \phi)$ in Eq. (3) should have been denoted as $\mathcal{R}_n^m(\rho, \phi)$, but the superscript $m$ is dropped for simplification. Considering $m$ is a good quantum number, doing so should not give rise to any confusion. Substituting Eq. (2) into the Schrödinger equation with the Hamiltonian (1), and projecting onto the basis $B_i \mathcal{R}_n$ yield the following matrix equation

$$HC = ENC ,$$

(4)

where the Hamiltonian matrix elements are of the form

$$H_{i', n': in} = -\frac{1}{2} \delta_{n', n} \int_0^{z_{\text{max}}} B_i(z) \frac{d^2}{dz^2} B_i(z) dz + n|\gamma| \int_0^{z_{\text{max}}} B_i(z) B_i(z) dz + \int_0^{z_{\text{max}}} B_i(z) V_{n', n}(z) B_i(z) dz ,$$

(5)

and $N$ is the overlap matrix with elements

$$N_{i', n': in} = \delta_{n', n} \int_0^{z_{\text{max}}} B_i(z) B_i(z) dz .$$

(6)

The upper limits of the integration, $z_{\text{max}}$, in Eqs. (5) and (6) denotes the maximum of coordinate $z$. It changes with energy levels and magnetic field strengths. $z_{\text{max}}$ should be taken to be large enough to obtain stable eigenvalues in calculations. The overlap matrix $N$ is a positive definite real symmetric matrix. Its existence is attributed to the B-spline basis nonorthogonality. The matrix element $V_{n', n}(z)$ in Eq. (5) is given by\textsuperscript{[21]}

$$V_{n', n}(z) = -\int_0^{\infty} \mathcal{R}_{n'}(\rho) \frac{1}{\sqrt{\rho^2 + z^2}} \mathcal{R}_n(\rho) \rho d\rho$$

$$= -\frac{2}{\sqrt{\pi}} \sum_{j=0}^{\text{min}(n', n)} \left[ \frac{n'!n!(n' + |m|)!(|n + |m||)!)}{(n' - j)!(n - j)!(|n + |m||)!j!} \right] \sqrt{\frac{\pi}{2}} \int_0^{x_{\text{max}}} (\sin x)^{2\beta} (\cos x)^{2\alpha}$$

$$\times \exp \left[-\frac{\gamma^2 \cos^2 x}{2 \sin^2 x} \right] dx .$$

(7)

The adaptive Gauss–Kronrod quadrature and the Gaussian quadrature are, respectively, used to evaluate the integral of Eq. (8) numerically, and all the matrix elements of $H$ and $N$. For states of even and odd parity, we perform calculations at the whole space of $z$ with $-z_{\text{max}} \leq
$z \leq z_{\text{max}}$ and the half space of $z$ with $0 \leq z \leq z_{\text{max}}$, respectively. The combination of such a choice and properties of B-spline functions make it easy to enforce boundary conditions. That is to simply remove $i = 1, N$ terms from the summation over $i$ in Eq. (2), namely $B_i(z)$ and $B_N(z)$, where $N$ represents the number of B-spline functions. Once the calculations of the matrix elements are finished, standard routines for matrix diagonalization can be used to obtain the eigenvalues and eigenvectors numerically.

It should be pointed out that this method is valid only for strong fields, because the basis size required to obtain stable convergence becomes larger and larger with decreasing field strengths. In the situation of weak fields, we will adopt the finite-basis-set technique,[16] in which the Schrödinger equation is solved in spherical coordinates by expanding wave functions in terms of a B-spline basis in the radial direction and spherical harmonics in the angular direction. The finite-basis-set technique was developed recently in order to calculate ground and lowly excited states for magnetized hydrogen atoms. Its details have been presented and therefore are omitted here. We will combine the current method and the finite-basis-set technique for computations of circular Rydberg states of atomic hydrogen for arbitrary magnetic field strengths.

3 Results and Discussion

Circular Rydberg states with $|m| = 10 - 70$ were calculated in a wide field strength range. Only the eigenvalue problem for states with negative $m$ is solved, since eigenvalues for states with positive $m$ are easily obtained by an energy shift from the corresponding states of negative $m$.[14] The maximum number and order of B-spline functions were taken to be $N = 100$ and $k = 11$, respectively. A crucial procedure in the current calculation is to suitably select the knot sequence of B-spline functions and maxima of space coordinates, $z_{\text{max}}$ and $r_{\text{max}}$, where $r_{\text{max}}$ represents the maximum of radial coordinates $r$. For calculations in cylindrical coordinates with the current method, $z = \{0, z_{\text{max}}\}$ was divided into the inner and outer region, and in each region an equidistant mesh was taken. Furthermore, the mesh size in the inner region was selected to be equal to twice the mesh size in the outer region. Such a choice is expected to characterize the behavior of circular electron motion in the combined Coulomb and magnetic fields quite well. For calculations in spherical coordinates with the finite-basis-set technique, an exponential mesh was adopted, following Ref. [16] (see this reference for details). Energy levels were optimized by adjusting the distribution of the knots, and $z_{\text{max}}$ (or $r_{\text{max}}$).

For any given state, there does exist a magnetic field range with strong competition between Coulomb and magnetic interactions. In this range, the solution of the Schrödinger equation is very difficult. Either a gap of fields, where energy levels can not be directly calculated because of the limitation of the used method, remain left or the gap is smoothly jointed to the results from the two sides of the gap (see, i.e., Ref. [14] and references therein). Our method, however, is very effective in such a field range. Table 1 lists eigenvalues for circular states with $m = -10$ and $-70$ in a magnetic field with strengths ranging from 0.0001 to 0.1 a.u., calculated in the current method and the finite-basis-set technique. The uncertainty of each eigenvalue is $\pm 1$ in the last digit, except for those in a range from 0.0001 to 0.002 a.u. in the second column and from 0.02 to 0.1 a.u. in the third column. The two methods are in excellent agreement.

The numbers of spherical harmonics and Landau states, which are used to expand wave functions, are also given in this table. The numbers reflect symmetry of the system at a given state and field strength because of the competition between Coulomb and magnetic interactions. For example, let us look at the circular state with $m = -10$. At $\gamma = 0.0001$ a.u., the system is more spherically symmetric (the number of spherical harmonics is 2), as the Coulomb interaction is dominant, while the magnetic interaction is relatively small, but at $\gamma = 0.1$ a.u., the symmetry is completely broken (the number of spherical harmonics and Landau states is close), as the two kinds of interactions become comparable. Now, let us turn to the $m = -70$ state. At $\gamma = 0.1$ a.u., the system is more cylindrically symmetric (the number of Landau states is 5), as the magnetic interaction is dominant, while the Coulomb interaction is relatively small, but at $\gamma = 0.0001$ a.u., the symmetry is completely broken, because the two interactions become comparable.

It is time saving to adopt the different method for systems with more spherical and more cylindrical symmetries. Totally, the method developed in this paper is not suitable for weak magnetic fields, while the finite-basis-set technique is not suitable for strong fields. However, both methods remain valid for intermediate fields, and should be in excellent agreement. Eigenvalues are calculated and listed in Table 2 for circular Rydberg states with $m$ from $-20$ to $-60$ in a magnetic field range from zero to $10^4$ a.u. We first use the finite-basis-set technique when the magnetic field is small and then switch to the current method when it becomes larger than some value.

We made a comparison between the present eigenvalues for the circular state with $m = -24$ with available theoretical data,[14–15,17] and excellent agreement is found. The compared data were not listed here as doing so is obviously superfluous. Restricted by the used spherical coordinate system, the method of Liu et al.[15] is ineffective for higher fields. One may see that their accuracy decreases with increasing field strengths. At $\gamma = 0.0003$ a.u., our energy agrees very well with the highly accurate result of Germann et al.[17] Our calculation gives $-5.315 899 713 035$ a.u. As previously mentioned, here we point out again that not in all field strengths circular states may be calculated with the dimensional perturbation theory of Germann et al. because of the limitation of this theory (also see Ref. [18]).
Table 1  Eigenvalues for circular Rydberg states with \( m = -10 \) and \(-70 \) in units of Hartree in an intermediate magnetic field strength, calculated with the finite-basis-set technique (Sph.) and the method developed in this paper (Lan.).

| \( \gamma \)/a.u. | \( m = -10 \) | \( m = -70 \) |
|------------------|--------------|--------------|
| \( 1.0 \)[-4]    | \(-5.622 \times 10^{-4} \) | \(-8.572 \times 10^{-4} \) |
| \( 2.0 \)[-4]    | \(-5.153 \times 10^{-4} \) | \(-1.197 \times 10^{-4} \) |
| \( 4.0 \)[-4]    | \(-6.034 \times 10^{-4} \) | \(-1.677 \times 10^{-4} \) |
| \( 6.0 \)[-4]    | \(-6.805 \times 10^{-4} \) | \(-2.044 \times 10^{-4} \) |
| \( 8.0 \)[-4]    | \(-7.493 \times 10^{-4} \) | \(-2.353 \times 10^{-4} \) |
| \( 1.0 \)[-3]    | \(-8.118 \times 10^{-4} \) | \(-2.624 \times 10^{-4} \) |
| \( 2.0 \)[-3]    | \(-1.065 \times 10^{-3} \) | \(-3.683 \times 10^{-4} \) |
| \( 4.0 \)[-3]    | \(-1.431 \times 10^{-3} \) | \(-5.170 \times 10^{-4} \) |
| \( 6.0 \)[-3]    | \(-1.712 \times 10^{-3} \) | \(-6.303 \times 10^{-4} \) |
| \( 8.0 \)[-3]    | \(-1.948 \times 10^{-3} \) | \(-7.254 \times 10^{-4} \) |
| \( 1.0 \)[-2]    | \(-2.155 \times 10^{-3} \) | \(-8.088 \times 10^{-4} \) |
| \( 2.0 \)[-2]    | \(-2.961 \times 10^{-3} \) | \(-1.133 \times 10^{-3} \) |
| \( 4.0 \)[-2]    | \(-4.080 \times 10^{-3} \) | \(-1.586 \times 10^{-3} \) |
| \( 6.0 \)[-2]    | \(-4.924 \times 10^{-3} \) | \(-1.930 \times 10^{-3} \) |
| \( 8.0 \)[-2]    | \(-5.627 \times 10^{-3} \) | \(-2.217 \times 10^{-3} \) |
| \( 1.0 \)[-1]    | \(-6.240 \times 10^{-3} \) | \(-2.469 \times 10^{-3} \) |

\(^a\)A\[B\] = \( A \times 10^B \); \(^b\)The number in parenthesis gives the number of spherical harmonics; \(^c\)The number in parenthesis gives the number of Landau states.

Table 2  Eigenvalues for circular Rydberg states with \( m \) from \(-20 \) to \(-60 \) in units of Hartree and as a function of magnetic field strengths ranging from \( \gamma = 0 \) to \( 10^4 \). The uncertainty of each eigenvalue is \( \pm 1 \) in the last digit.

| \( \gamma \) | \( m = -20 \) | \( m = -30 \) | \( m = -40 \) | \( m = -50 \) | \( m = -60 \) |
|-------------|--------------|--------------|--------------|--------------|--------------|
| 0           | \(-1.133 \times 10^{-4} \) | \(-1.000 \times 10^{-4} \) | \(-9.000 \times 10^{-4} \) | \(-8.118 \times 10^{-4} \) | \(-7.317 \times 10^{-4} \) |
| 1.0[-6]     | \(-9.000 \times 10^{-4} \) | \(-8.118 \times 10^{-4} \) | \(-7.317 \times 10^{-4} \) | \(-6.805 \times 10^{-4} \) | \(-6.240 \times 10^{-4} \) |
| 1.0[-5]     | \(-8.118 \times 10^{-4} \) | \(-7.317 \times 10^{-4} \) | \(-6.805 \times 10^{-4} \) | \(-6.240 \times 10^{-4} \) | \(-5.707 \times 10^{-4} \) |
| 1.0[-4]     | \(-7.317 \times 10^{-4} \) | \(-6.805 \times 10^{-4} \) | \(-6.240 \times 10^{-4} \) | \(-5.707 \times 10^{-4} \) | \(-5.170 \times 10^{-4} \) |
| 1.0[-3]     | \(-6.805 \times 10^{-4} \) | \(-6.240 \times 10^{-4} \) | \(-5.707 \times 10^{-4} \) | \(-5.170 \times 10^{-4} \) | \(-4.696 \times 10^{-4} \) |
| 1.0[-2]     | \(-6.240 \times 10^{-4} \) | \(-5.707 \times 10^{-4} \) | \(-5.170 \times 10^{-4} \) | \(-4.696 \times 10^{-4} \) | \(-4.276 \times 10^{-4} \) |
| 1.0[-1]     | \(-5.707 \times 10^{-4} \) | \(-5.170 \times 10^{-4} \) | \(-4.696 \times 10^{-4} \) | \(-4.276 \times 10^{-4} \) | \(-3.806 \times 10^{-4} \) |
| 1.0[+1]     | \(-4.276 \times 10^{-4} \) | \(-3.806 \times 10^{-4} \) | \(-3.228 \times 10^{-4} \) | \(-2.754 \times 10^{-4} \) | \(-2.276 \times 10^{-4} \) |
| 1.0[+2]     | \(-3.228 \times 10^{-4} \) | \(-2.754 \times 10^{-4} \) | \(-2.276 \times 10^{-4} \) | \(-1.930 \times 10^{-4} \) | \(-1.560 \times 10^{-4} \) |
| 1.0[+3]     | \(-2.276 \times 10^{-4} \) | \(-1.930 \times 10^{-4} \) | \(-1.560 \times 10^{-4} \) | \(-1.276 \times 10^{-4} \) | \(-1.000 \times 10^{-4} \) |

\(^a\)A\[B\] = \( A \times 10^B \).
Intermediate electron clouds are separated because the magnetic interaction is becoming significant. At $\gamma = 0.1$, the Lorenz force is dominant. To reach the same convergence as 16 Landau states, we contained the 38 spherical partial waves.

![Contour plots of probability densities](image1)

Fig. 1 (Color online) Contour plots of probability densities of the electron in a circular state with $m = -24$, integrated over the angular variable of cylindrical coordinates ($\rho, z, \phi$). The magnetic field is along the $z$-axis. The total wave function is normalized such that $\int |\Psi(\rho, z, \phi)|^2 \rho d\rho dz d\phi = 1$. Note: The data aspect ratio is taken to be 1:1 between $z$ and $\rho$.

![Probability densities](image2)

Fig. 2 (Color online) Probability densities of the electron in a non-circular state $7g_{-4}$, integrated over the angular variable of cylindrical coordinates ($\rho, z, \phi$). The direction of magnetic fields and the normalization of wave function are the same as those in Fig. 1.
4 Summary and Conclusion

A theoretical scheme using a B-spline basis set has been proposed to compute circular Rydberg states of hydrogen atoms in the intermediate magnetic field. We solved the Schrödinger equation in the spherical and cylindrical coordinate, corresponding to weak and strong magnetic fields, respectively. The scheme is valid for an arbitrary magnetic field, and can produce high accuracy energy levels (Note: the high accuracy here indicates that in the theoretical framework defined). We showed how to save computational time as well as to obtain a high accuracy. The energy levels of atomic hydrogen are presented for circular Rydberg states with azimuthal quantum numbers $|m| = 10−70$ as a function of magnetic field strengths ranging from zero to $2.35 \times 10^9$ T. The competition between Coulomb and magnetic interactions is discussed and illustrated.

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