Fine structure in a strong magnetic field: Paschen-Back effect reconsidered in Rydberg atoms

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Using a kind of potential model wave function for alkali metal atoms, we nonperturbatively study the effect of fine structure on the Rydberg spectra of Cs atom in a strong magnetic field. Our numerical results reveal spectral structure dramatically different from the well-established Paschen-Back effect, and we argue that the fine structure of the Rydberg Cs atom cannot be neglected even in a magnetic field as strong as several teslas. We also give an error estimate of our results and a word on possible experimental verification.

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INTRODUCTION

Until now there have been numerous theoretical and experimental studies of Rydberg states in external fields from the viewpoints of both energy spectra and wave packet dynamics. To our knowledge there have been no efforts concentrated on the spin-orbit interaction induced fine structure (FS) effects of the Rydberg atom in external fields, except one experimental work which concerns the Stark structure of the Cs atom [1] and one theoretical work of our own which concerns the Stark evolution of the diamagnetic structure of the Cs atom [2]. Such negligence of the FS effect in Rydberg states has occurred for two reasons. First, as principal quantum number n increases, FS splittings scale down as n^{-3}, in sharp contrast to the rapid increase of Stark ($\sim n^2$) and quadratic Zeeman ($\sim n^4$) interactions. Therefore FS effects are believed to be of perturbative magnitude compared with the effects of external fields in the Rydberg situation. Second, as has been shown in many textbooks [3–5], a strong enough magnetic field decouples the spin-orbit interaction and leads to what is usually termed the Paschen-Back (PB) effect (or complete Paschen-Back effect, according to some authors), which indicates that spin-related properties can be omitted in spectroscopic studies when the magnetic field is strong enough.

In this paper we investigate the properties related to the spin-orbit interaction of a heavy atom such as Cs, which has a large FS effect, in an external magnetic field. We show that the evolution of a FS doublet is dramatically different from the monotonic PB effect and the FS effect of Rydberg states can be observed even in the presence of a very strong magnetic field. In this sense one cannot rely comfortably on the well-established PB effect to completely ignore the spin-orbit coupling of Rydberg atoms in a strong magnetic field.

METHOD

A multiparameter model for the core potential of an alkali metal atom was proposed by He *et al.* [6]. From this potential model the FS radial wave functions R_{nlj}, which have a multiparameter analytical form, were deduced with experimental energies E_{nlj} as the only input parameters [2]. The total wave functions of the field-free alkali metal atom are $\Psi_{nljm_j}=R_{nlj}\phi_{ljm_j}$, where ϕ_{ljm_j} is the LS-coupled angular wave function. Such potential model (PM) wave functions have been successfully used in many cases [2,7]. Using PM wave functions as bases, we diagonalize the following Hamiltonian to obtain the eigenstates and hence the spectroscopic properties in an external magnetic field (magnetic field B is in the z axis and we use atomic units):

$$H=H_0+\gamma(m_j+S_z)/2+\gamma^2r^2\sin^2\theta/8, \tag{1}$$

where H_0 is the field-free Hamiltonian such that $H_0\Psi_{nljm_j}=E_{nlj}\Psi_{nljm_j}$. m_j is a good quantum number for $J_z=L_z+S_z$, and $\gamma=B/B_0$ is magnetic field strength measured in atomic unit $B_0=2.35\times10^5$ T. In this way the FS is nonperturbatively included. Although such PM radial wave functions are not orthogonal due to the state dependence of the potential model parameters, the overlaps $\langle R_{nlj}|R_{n'lj}\rangle$ for the Rydberg states of our concern are very small (typically 10^{-5} compared to unity for $n>20$), since the parameters vary very little from one state to another for fixed l,j and $n>20$. So we do not have to deal with generalized eigenvalue problems.

PASCHEN-BACK EFFECT

The evolution of a single FS doublet with magnetic field can be approximately obtained by diagonalizing the following matrix of Hamiltonian Eq. (1) in the 2×2 subspace spanned by zero field eigenstates $\Psi_{nlj_-m_j}$ and $\Psi_{nlj_+m_j}$, $j_\pm=l\pm1/2$:

TABLE I. The evolution of $27P$ FS doublet ($m_j=1/2$) with magnetic field. B is the magnetic field in tesla. ΔE is the wave number (cm^{-1}) of the laser for excitation from $15S$ state. f is the oscillator strength. $\langle S_z\rangle$ is the expectation value of S_z. Entry marks: PB; Paschen-Back argument; PM, our potential model basis method; I, lower level of the doublet; II, upper level of the doublet. Numbers in brackets indicate powers of 10.

B		0.5	1.0	1.5	2.0	2.5	3.0	3.5	4.0	4.5	5.0
PB.I	ΔE	715.991	714.792	714.579	714.359	714.134	713.908	713.679	713.450	713.220	712.989
	f	3.04[-5]	1.78[-5]	1.12[-5]	7.61[-6]	5.45[-6]	4.07[-6]	3.14[-6]	2.50[-6]	2.03[-6]	1.68[-6]
	$\langle S_z\rangle$	-0.312	-0.389	-0.430	-0.452	-0.466	-0.474	-0.480	-0.484	-0.487	-0.489
PB.II	ΔE	715.661	715.628	715.607	715.594	715.585	715.578	715.573	715.568	715.565	715.563
	f	1.31[-4]	1.44[-4]	1.50[-4]	1.54[-4]	1.56[-4]	1.58[-4]	1.58[-4]	1.59[-4]	1.60[-4]	1.60[-4]
	$\langle S_z\rangle$	0.312	0.389	0.430	0.452	0.466	0.474	0.480	0.484	0.487	0.489
PM.I	ΔE	715.059	715.070	715.215	715.496	715.909	716.442	717.066	717.733	718.419	719.146
	f	1.61[-5]	1.06[-5]	9.05[-6]	9.88[-6]	1.36[-5]	2.36[-5]	4.65[-5]	7.95[-5]	9.90[-5]	1.02[-4]
	$\langle S_z\rangle$	-0.293	-0.345	-0.361	-0.353	-0.316	-0.226	-0.033	0.225	0.386	0.451
PM.II	ΔE	715.706	715.797	715.976	716.239	716.583	717.025	717.580	718.292	719.175	720.194
	f	1.23[-4]	1.28[-4]	1.30[-4]	1.28[-4]	1.24[-4]	1.12[-4]	8.73[-5]	5.04[-5]	2.45[-5]	1.27[-5]
	$\langle S_z\rangle$	0.293	0.345	0.361	0.353	0.316	0.226	0.033	-0.225	-0.386	-0.451

$$\begin{pmatrix} E_{nlj_-}+\frac{\gamma}{2}\left(\frac{m_j}{2}-\frac{m_j}{2l+1}\right)+\frac{\gamma^2}{8}D_{11} & -\frac{[(l+1/2)^2-m_j^2]^{1/2}}{2l+1}\gamma O_{nl}+\frac{\gamma^2}{8}D_{12} \\ -\frac{[(l+1/2)^2-m_j^2]^{1/2}}{2l+1}\gamma O_{nl}+\frac{\gamma^2}{8}D_{12} & E_{nlj_+}+\frac{\gamma}{2}\left(\frac{m_j}{2}+\frac{m_j}{2l+1}\right)+\frac{\gamma^2}{8}D_{22} \end{pmatrix}, \tag{2}$$

where

$$O_{nl}=\int R_{nlj_-}R_{nlj_+}r^2dr, \quad D_{11}=\int \Psi_{nlj_-m_j}r^2\sin^2\theta\Psi_{nlj_-m_j}d\tau,$$

$$D_{22}=\int \Psi_{nlj_+m_j}r^2\sin^2\theta\Psi_{nlj_+m_j}d\tau, \quad D_{12}=\int \Psi_{nlj_-m_j}r^2\sin^2\theta\Psi_{nlj_+m_j}d\tau.$$

In what we call the PB argument (see, e.g., Ref. [3]), both R_{nlj_-} and R_{nlj_+} are approximated with R_{nl}, hence $O_{nl}=1$, and the quadratic B^2 terms are omitted for low excited states; the eigenenergies of the above matrix thus simplified can then be explicitly written as

$$E_{nl_\pm}=\frac{1}{2}(E_{nlj_-}+E_{nlj+})+\frac{\gamma}{2}m_j \pm\left(\frac{1}{4}\Delta E_{fs}+\frac{\gamma}{2}\frac{m_j}{2l+1}\Delta E_{fs}+\frac{\gamma^2}{16}\right)^{1/2}, \tag{3}$$

where $\Delta E_{fs}=E_{nlj_+}-E_{nlj_-}$. If $\gamma/\Delta E_{fs}\gg 1$, the two FS levels $E_{nl_\pm}\to\frac{1}{2}(E_{nlj_-}+E_{nlj_+})+(\gamma/2)m_j\pm\frac{1}{2}(\gamma/2)$ and the corresponding wave functions reduce to LS-decoupled spin-up $R_{nl}Y_{l,m_j-1/2}\binom{1}{0}$ and spin-down $R_{nl}Y_{l,m_j+1/2}\binom{0}{1}$, respectively, as if the FS no longer exists and S_z becomes a constant of motion as in the case of S states. This is the PB effect caused by the decoupling of spin-orbit interaction by a magnetic field. If in a given m_j space we denote with $\downarrow$ ($\uparrow$) an initial spin-down (-up) S state and denote with $\Downarrow$ ($\Uparrow$) a final spin-down (-up) Rydberg state in the PB regime, the dipole excitations from $\downarrow$ to $\Downarrow$, denoted as $\downarrow\Downarrow$ transitions, give identical spectral levels with the corresponding $\uparrow\Uparrow$ transitions, because in a given m_j space the decoupled spin-down subspace ($\downarrow$ and $\Downarrow$ states) and spin-up subspaces ($\uparrow$ and $\Uparrow$ states) are identical except for a global S_z-induced paramagnetic energy shift $\gamma/2$. The $\downarrow\Uparrow$ and $\uparrow\Downarrow$ transitions have vanishing intensities because $\Delta m_s=0$ for dipole transitions. Therefore the degree of freedom of spin is not detectable in spectral observation; in other words FS can be neglected in the PB regime.

The evolution to the PB regime in $m_j=1/2$ subspace ($\uparrow\Downarrow$, $\uparrow\Uparrow$ transitions) is numerically manifested in entry PB of Table I, where we tabulate the $27P$ doublet with respect to the transitional initial state $15S\uparrow$. The lower (upper) level is denoted by I (II). We see that as B increases the I and II states are gradually decoupled: $\uparrow\Uparrow$ levels take up ever more dominant intensities, and in the meantime each state gains more and more definite spin identities, viz., $\langle S_z\rangle\to-1/2$ for I states and $\langle S_z\rangle\to 1/2$ for II states. Therefore, the I (II) state corresponds asymptotically to $\uparrow\Downarrow$ ($\uparrow\Uparrow$) level as the PB effect builds up.

Where Rydberg states of Cs are concerned, the FS splittings are so large (e.g., $\Delta E_{fs}\sim 0.5\ \mathrm{cm}^{-1}$ for $27P$) that no magnetic field can be on the one hand strong enough to decouple the spin-orbit interaction and on the other hand weak enough to make the B^2 term negligible. So the PB

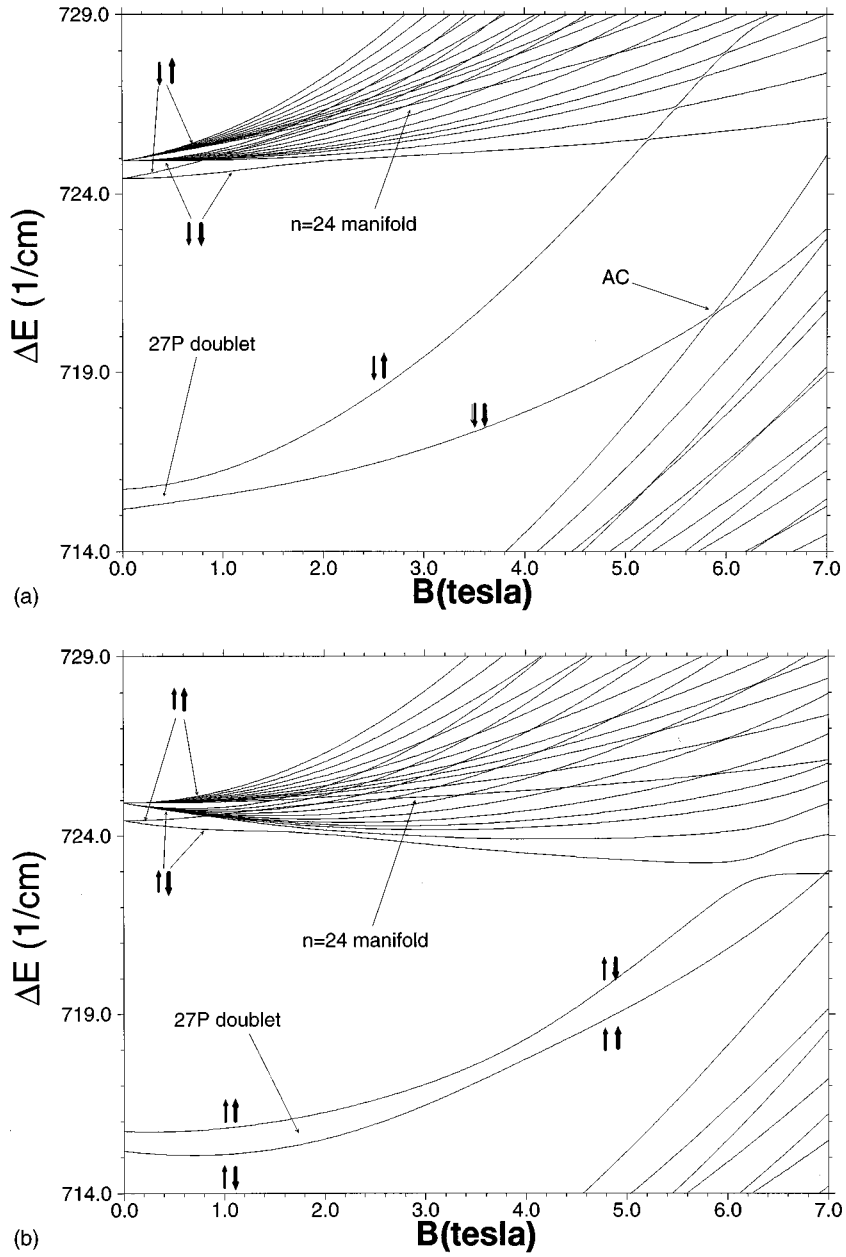

FIG. 1. (a) The spectral lines π-excited from $15S$, $m_s=-1/2$ state. ΔE is the level energy with respect to the initial state. (b) The spectral lines π-excited from $15S$, $m_s=1/2$ state. ΔE is the level energy with respect to the initial state. (c) The superposition of (a) and (b).

argument does not apply here and numerical calculations, which nonperturbatively include the FS effect and the diamagnetic B^2 term, are inevitable. Our PM method offers an alternative.

RESULTS

Our objective is to give numerically simulated spectra excited from a spin unpolarized beam of Cs atom ($^2S_{1/2}, m_j=m_s=\pm 1/2$). Being concerned only with the FS of high Rydberg states, we select $15S$, instead of $6S$, as the initial state of excitation to higher Rydberg states, since it has a very small hyperfine structure besides being free of FS. The results of our PM method for the $27P$ FS doublet are shown in Fig. 1 and entry PM of Table I. The tabulated numerical results are converged with respect to the enlargement of the basis set and the linear and quadratic effects of the magnetic field on the initial $15S$ state are also taken into account. We find that the quadratic term's contributions to the energy of the $15S$ state are almost identical (to within 0.0001 cm^{-1}) in the two $m_j=m_s=\pm 1/2$ subspaces, so the quadratic field effect of the initial state does not cause any complication of the Rydberg spectra revealing the FS features of interest to us.

The behavior of the $n=24$ manifolds (levels composed mainly of $l\geqslant 3$ components shown in the figures as the level bundles) in both $m_j=-1/2$ and $m_j=1/2$ spaces are trivially simple. They have very small FS and the onset of a small magnetic field immediately decouples them into spin-up and spin-down groups. $\downarrow\Downarrow$ and $\uparrow\Uparrow$ levels coincide perfectly, as can be seen in Fig. 1(c) and our numerical results show that $\downarrow\Uparrow$ and $\uparrow\Downarrow$ levels have oscillator strengths one or two orders of magnitudes lower than those of $\downarrow\Downarrow$ and $\uparrow\Uparrow$.

The most significant feature, which appears in the $m_j=1/2$ space [Fig. 1(b)], is what we call the ''gradual spin flip.'' In entry PM of Table I we see that as B increases up to

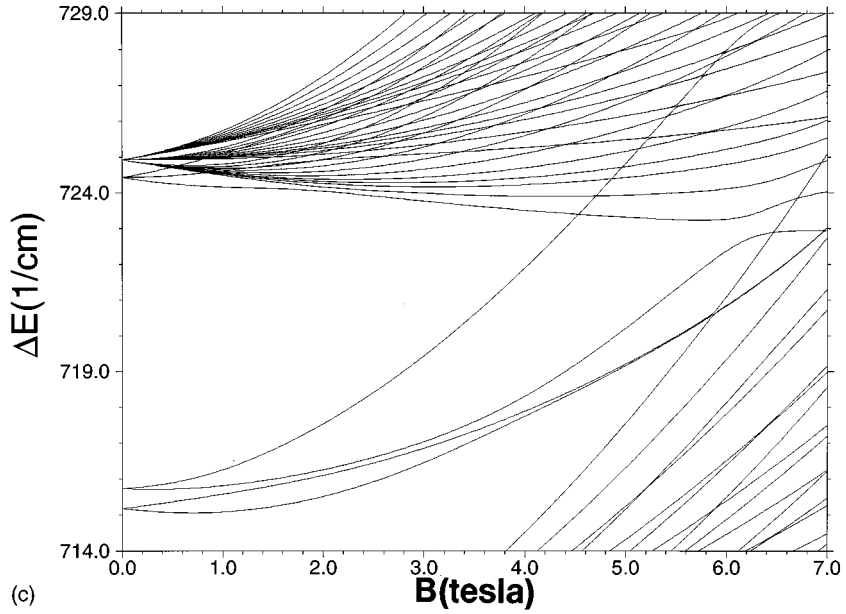

FIG. 1 (*Continued*).

$B \simeq 1.5$ T, the upper (lower) level of the $27P$ doublet gains spin-up (-down) character in accordance with the PB effect. But from $B \simeq 1.5$ T onward, the evolutions of the features of the two levels are reversed, viz., for the upper (lower) level, oscillator strength goes down (up) and $\langle S_z \rangle$ goes down (up) to zero and then negative (positive). If we adiabatically trace either level we see a gradual spin flip. Around $B \simeq 3.5$ T the spacing between the two levels reaches a minimum and their values of $\langle S_z \rangle$ are nearly zero and oscillator strengths are almost equally shared among the two levels. In a trial diagonalization with only $\Psi_{nlj_+m_j}, \Psi_{nlj_-m_j}$, i.e., the diagonalization of matrix 2, we also find such a spin flip but at a lower B (~ 0.5 T). It is accompanied by the sign flip (from negative to positive) of the off diagonal element $-(\{[(l+1/2)^2 - m_j^2]^{1/2}\}/(2l+1))\gamma O_{nl} + (\gamma^2/8) D_{12}$, as the quadratic term $(\gamma^2/8) D_{12}$, which is positive for $m_j = 1/2$, gradually turns dominant. It turns out that the enlargement of the basis set the to the PM basis only ''delays'' the spin flip to a larger B. So we can say that the spin flip is due to the quadratic effect of the magnetic field, especially the term $(\gamma^2/8) D_{12}$. In fact, such a spin flip can be regarded as an avoided crossing (AC) between two levels which otherwise belong to two approximately decoupled m_s subspaces in a given m_j space. In the vicinity of the AC both levels are deprived of any distinct spin identity due to the inter-m_s coupling induced by strong LS interaction. After the AC the two levels exchange their features (and hence their denotations) and the decoupling process, which can be called a quadratic PB effect, resumes. The overall process can be sketched as follows in terms of $|m_l m_s\rangle$

Zero field:

$$\text{Upper level:} \quad \sim \sqrt{2/3}|0+\rangle - \sqrt{1/3}|1-\rangle,$$

$$\text{Lower level:} \quad \sim \sqrt{1/3}|0+\rangle + \sqrt{2/3}|1-\rangle.$$

Linear PB regime:

$$\text{Upper level:} \quad \sim |0+\rangle,$$

$$\text{Lower level:} \quad \sim |1-\rangle.$$

Spin flipping regime:

$$\text{Upper level:} \quad \sim \sqrt{1-\alpha^2}|0+\rangle - \alpha|1-\rangle, \quad 0 < \alpha < 1 \text{ and } \alpha \text{ increases with } B,$$

$$\text{Lower level:} \quad \sim \sqrt{1-\beta^2}|0+\rangle + \beta|1-\rangle, \quad 0 < \beta < 1 \text{ and } \beta \text{ decreases with } B.$$

Quadratic PB regime:

$$\text{Upper level:} \quad \sim |1-\rangle,$$

$$\text{Lower level:} \quad \sim |0+\rangle.$$

The behavior of the $27P$ doublet in $m_j=-1/2$ space, however, is qualitatively the same as the PB effect, as shown in Fig. 1(a). So its numerical results are not tabulated. From $B=5$ T onward in the depicted field range in Fig. 1(c), $\downarrow\Downarrow$ and $\uparrow\Uparrow$ levels are quasidegenerate except for a small spacing of the order 0.001 cm^{-1} which requires high resolution to be resolvable. This is the quadratic PB regime. But even in this regime an important phenomenon caused by the FS effect also appears in the $m_j=-1/2$ space, viz., the avoided crossing between the lower level of the $27P$ doublet (an $\Downarrow$ state) and the highest $\Uparrow$ level of the $n=23$ manifold lying below, marked as AC in Fig. 1(a) at $B\simeq 5.87$ T. It reflects the residual FS effect which takes significant effect at local zones where levels of different decoupled m_s subspaces in an m_j space are encountered. The inter-m_s coupling is manifested in the considerable AC spacing of 0.06 cm^{-1} between the two levels, their comparable oscillator strengths (3.8×10^{-5} for the lower and 4.9×10^{-5} for the upper level), and their vanishingly small $\langle S_z\rangle$ values (~ 0.04). From Fig. 1(c), and more clearly from numerical results not listed here, we see that at this AC the $\uparrow\Uparrow$ level, which is quasidegenerate with $\downarrow\Downarrow$, lies nearly halfway between the two avoided-crossing levels and has a larger oscillator strength (8.5×10^{-5}). Therefore a spectrum with a resolution of about 0.01 cm^{-1} may reveal such an interesting sign of the residual FS effect in a strong PB regime. In principle such inter-m_s AC's appear wherever $\Downarrow$ and $\Uparrow$ levels are encountered, though they may be unresolvable or have too low oscillator strengths.

The existence of the wide-spanning spin flip phenomena and the local inter-m_s AC discussed above show that the well-established PB effect is dramatically modified by the quadratic field term and the effect of FS on the Rydberg spectra cannot be neglected even in such a strong field as several teslas. Our results highlight an important aspect of the spin-related features in the angular momentum couplings of different schemes (LS-, JJ-coupling) in many heavy atoms.

ERROR ESTIMATE

The phenomena numerically observed here are the results of the competition among such different factors as the linear, quadratic field effects and the FS effect. At what field strength the phenomena appear, or whether they appear at all, is dependent on the relative magnitudes of these effects. So it is indispensible that we give an error estimate of our results so as to exclude the possibility of artifact due to numerical inaccuracy.

Let $H=\{h_{ij}\}_{i,j=1}^{N}$, $\Delta H=\{\Delta h_{ij}\}_{i,j=1}^{N}$, and $H+\Delta H=\{h_{ij}+\Delta h_{ij}\}_{i,j=1}^{N}$ be three real symmetric matrices which have eigenvalues $\{\epsilon_i\}$, $\{\delta_i\}$, and $\{\epsilon_i+\Delta\epsilon_i\}$, respectively; it can be proven that the following inequality holds (Hoffman-Wielandt Theorem [8]):

$$\overline{\Delta\epsilon}\equiv\left(\frac{1}{N}\sum_{i=1}^{N}(\Delta\epsilon_i)^2\right)^{1/2}\leqslant\left(\frac{1}{N}\sum_{i=1}^{N}(\delta_i)^2\right)^{1/2}=\left(\frac{1}{N}\sum_{i,j=1}^{N}(\Delta h_{i,j})^2\right)^{1/2}. \tag{4}$$

Define $\Delta h_{\max}=\max|\Delta h_{ij}|$ and suppose both H and $H+\Delta H$ are banded matrices with full bandwidth M; we readily get from this inequality

$$\overline{\Delta\epsilon}\leqslant M^{1/2}\Delta h_{\max}. \tag{5}$$

Regardless of the particular subroutines adopted in our code for diagonalizations, the above inequality applies to our Hamiltonian matrix and can be used for the error estimate of the eigenenergies, where $\overline{\Delta\epsilon}$ is the root mean square (rms) of errors of energies and Δh_{ij} are the errors of the elements of the Hamiltonian matrix. From Eq. (1) we see that errors Δh_{ij} have two sources: one is the experimental energies E_{nlj}; the other is the matrix elements of r^2 in our PM basis. For E_{nlj} we collected high-resolution experimental results from several groups [9–11] or used the fitted formula provided therein. The accuracies for the energies of Rydberg states of interest to us are generally better than 0.000 15 cm^{-1}. Given the sufficiently dense mesh and quadruple precision (32 significant decimal digits) used for numerical integration, the errors of r^2 matrix elements in the PM basis come not from numerical integration but from the inaccuracy of the potential model itself. Based on our past works (see Ref. [7]) and experiences, we are confident that our PM wave functions can give matrix elements of r^2 with relative error on the order of 10^{-3}. The magnitude of r^2 is on the order of n^4, e.g., $\langle 27P1/2|r^2|27P1/2\rangle=750\,745$ (a.u), and the error from the $\gamma^2r^2\sin^2\theta/8$ term is on the order of 10^{-3} cm^{-1} with the magnetic field and energy range of our concern. Therefore the errors of the Hamiltonian matrix elements come predominantly from the quadratic field term. The bandwith M of the Hamiltonian is about $n_{\max}$. $n_{\max}$ is the largest principal quantum number of the basis wave functions required for converged results and $n_{\max}<40$. Therefore we conclude from the preceding inequality that the average rms error of energy level is on the order of 0.01 cm^{-1}. To actually see the error of a particular level instead of the average, we adopt a Monte Carlo approach by diagonalizing the Hamiltonian with the ''key'' matrix elements (i.e., those of O_{nl} and r^2 for 27P1/2,3/2) randomly varying relatively between $1+0.5\%$ and $1-0.5\%$. The results show that the maximal errors thus observed are of the same order of magnitude as the average ones. But the actual probability is that random errors of matrix elements tend to cancel each other and the real error may be smaller than our estimates. Combining the average and the Monte Carlo estimates, we can say that our numerical representation of the spin flip phenomenon is accurate and, with less confidence, so is that for the inter-m_s AC.

EXPERIMENT

To simplify theoretical study, an S state, which has no FS and negligible hyperfine structure (HFS), is the preferred candidate for the initial state. In particular, the HFS splittings of such an S state should be less than 0.01 cm^{-1}. In the field range of our concern, which is well in the Paschen-Back regime of HFS, the remaining field independent HFS splitting [5] is $\delta_{hfs}=\alpha m_i m_j$, which is 0.001 75 cm^{-1} for $15S$. That is why we select $15S$ for theoretical study. But it causes difficulties for experimentalists because even though $15S$ can be sufficiently populated by two-

photon excitation from 6*S*, further excitations to higher Rydberg states may prove difficult due to the lack of suitable tunable infrared light sources. Since our theoretical conclusions remain the same for 6*S*, direct excitations from 6*S* to higher Rydberg states may be an plausible alternative as long as the overlapping energy levels arising from the HFS of the 6*S* state can be identified.

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