Vol 18 No 11, November 2009
1674-1056/2009/18(11)/4791-06

Chinese Physics B

© 2009 Chin. Phys. Soc.
and IOP Publishing Ltd

Scaled-energy spectroscopy of a $|M| = 1$ Rydberg barium atom in an electric field*

Wang Lei(汪 磊), Quan Wei(全 威), Shen Li(沈 礼),
Yang Hai-Feng(杨海峰), Shi Ting-Yun(史庭云), Liu Xiao-Jun(柳晓军),
Liu Hong-Ping(刘红平)†, and Zhan Ming-Sheng(詹明生)

State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071*, China*

(Received 13 March 2009; revised manuscript received 10 April 2009)

We observe strong energy-dependent quantum defects in the scaled-energy Stark spectra for $|M| = 1$ Rydberg states of barium atoms at three scaled energies: $\varepsilon = -2.000$, $\varepsilon = -2.500$ and $\varepsilon = -3.000$. In an attempt to explain the observations, theoretical calculations of closed orbit theory based on a model potential including core effect are performed for non-hydrogenic atoms. While such a potential has been uniformly successful for alkali atoms with a single valence electron, it fails to match experimental results for barium atoms in the 6snp Rydberg states with two valence electrons. Our study points out that this discrepancy is due to the strong perturbation from the 5d8p state, which voids the simple approximation for constant quantum defects of principle quantum number n.

Keywords: Stark, Rydberg, barium
PACC: 3150, 3260s, 3270

1. Introduction

The study of atoms inside external static magnetic and/or electric fields is of fundamental importance. An important tool for such studies is scaled-energy spectroscopy. The associated techniques and theories have been well developed over the years from many successful investigations of hydrogen and hydrogen-like alkali atoms with single valence electrons. Following closed orbit theory for atoms in external fields,[1−6] model potentials parameterized by quantum defects including core effects are introduced, leading to closed orbits analogous to those found in atomic hydrogen.[7] Quantum defect theory for single valence electron alkali atoms has shown great success as evidenced by nearly perfect agreement between theory and experiment.[7]

Despite this success, however, significant challenges remain especially for complex atoms with more than one valence electron. For example, for both alkaline-earth atoms barium[8,9] and strontium[10,11] with two valence electrons and a heavy core, theoretical calculations do not agree with the experimental scaled-energy spectra for the single excited Rydberg electron even after quantum defects are included to account for the effects of the core and the other valence electron. Adopting similar model potential formulations for alkaline-earth atoms, we found that although the peak positions of the experimental recurrence spectra agree with the closed orbit theory calculation, the peak intensities generally cannot be reproduced.[12] This is likely due to the fact that atoms with multi-valence electrons usually display considerably different quantum defects for different principle quantum numbers with excitation energy.[13] The changing quantum defect signals the failure of the simple model potential approach for barium[8,9] and strontium[10,11] atoms, despite the success in closed orbit theory calculations for hydrogen-like alkali Rydberg atoms in external fields. This sufficiently explains all experimental features of the scaled-energy spectra of sodium[14] and lithium,[15,16] as well as for the spectrum of helium Rydberg atoms in the narrow energy range.[17−21]

For the barium atom, experiments which focus exclusively on the configuration interaction in Stark dynamics within a multi-electron atom have been made to study the deviation from the standard one-electron picture.[22] The barium is excited by a direct laser to the 5d7d perturber which is embedded in and coupled to the single excited 6snd Rydberg series. A longer Kepler period than that observed in single-

*Project supported by the National Natural Science Foundation of China (Grant No 10774162) and the National Key Basic Research Program of China (Grant No 2005CB724501).
†Corresponding author. E-mail: liuhongping@wipm.ac.cn

electron systems at the same binding energy was identified. The dynamical effects associated with electron-electron interaction are identified by directly comparing and contrasting the scaled-energy recurrence spectra obtained from different energy windows.[23]

In our case, we focus on the effect of n-dependent quantum defects in the atoms of complex electron configuration in an external field. The 6snp Rydberg of barium are strongly perturbed by the 5d8p state and the quantum defect for the different main quantum number n varies dramatically.[24] The scaled-energy spectroscopy for excitation of barium atoms from $6s^2$ to 6snp provides a convenient probe for studying this interesting phenomenon of quantum defects that must analogously occur in strontium.[10] Furthermore, a better model potential can be developed to better carry out theoretical calculations to study the large core effect on the closed orbit, instead of simply using the hydrogenic Coulomb potential.[10] In previous studies, the resolution for laser wavelength and electric field in the scaled-energy spectroscopy for strontium[10] was not high enough to extract accurate information on the effect due to two valence electrons. A more reliable experiment is therefore urgently needed.

In this paper, we report an experimental observation of the scaled-energy spectra of barium at several scaled energies and the corresponding theoretical calculations for the recurrence spectra with closed orbit theory based on the hydrogen potential and model potentials. The experiment is performed by changing the electric field strength and the photon energy simultaneously while keeping the scaled energy $\varepsilon = E/F^{-1/4}$ constant. A configuration of one-photon excitation from the ground state is used to constrain the transitions to occur only from $6s^2$ to 6snp. The spectra are recorded at three scaled energies: $\varepsilon = -2.000$, -2.500 and -3.000, and the corresponding recurrence spectra are obtained by Fourier transformations. Two closed orbit theoretical calculations are performed. One is based on the hydrogenic pure Coulomb potential. The other employs the model potential widely used in alkali atoms with quantum defects. With the uncertainty of the scaled energy taken into account by a cutoff function,[25] we fail to reproduce the spectrum, especially for the strength of the spectral lines. This failure is due to the fact that barium shows a strong dependence of quantum defect on the excitation energy and this causes the core effects at different energies to be different. This observation also explains our previous work for strontium[10,11] and the work for barium by Kips, Kuik *et al.*[8,9]

2. Theory

The Hamiltonian of a barium Rydberg atom of energy E in an external electric field along the z axis is approximately given by

$$H = \frac{1}{2}\left(p_z^2 + p_\rho^2 + \frac{l_z^2}{\rho^2}\right) - \frac{1}{\sqrt{\rho^2+z^2}} + Fz + V_{\rm c}(z), \quad (1)$$

where $V_{\rm c}(r)$ is the centrical potential of the atomic core including the quantum defects. It describes short distance modifications of the ionic core when complete screening is not achieved. We employ a core potential in the following form:[7]

$$V_{\rm c}(r) = -\frac{1}{r}\left[1 + (Z-1)\,{\rm e}^{-a_1 r} + a_2 r {\rm e}^{-a_3 r}\right], \quad (2)$$

where the nuclear charges Z is 56 for barium and a_1, a_2 and a_3 are chosen to give the best agreement with the measured quantum defects. $a_1 = 4.987887$, $a_2 = 8.531785$ and $a_3 = 0.807332$ yield quantum defects $\mu_0 = 4.25183$, $\mu_1 = 3.80459$ and $\mu_2 = 2.75886$, which are close to the actual values $\mu_0 = 4.285$, $\mu_1 = 3.80$ and $\mu_2 = 2.76$.[9]

After introducing transformation

$$\hat{\rho} = \rho F^{1/2}, \quad \hat{z} = zF^{1/2}, \quad (3)$$

$$\hat{P}_\rho = P_\rho F^{-1/4}, \quad \hat{P}_z = P_z F^{-1/4}, \quad (4)$$

$$\hat{t} = tF^{1/4}, \quad (5)$$

the Hamiltonian becomes

$$\begin{aligned}\hat{H} &= HF^{-1/2} \\ &= \frac{1}{2}\left(\hat{P}_\rho^2 + \hat{P}_z^2 + \frac{\hat{L}_z^2}{\hat{\rho}^2}\right) \\ &\quad - \frac{1}{\sqrt{\hat{\rho}^2+\hat{z}^2}} + \hat{z} + V_{\rm c}(\hat{r}) \end{aligned} \quad (6)$$

with $\hat{L}_z = l_z F^{1/4}$. The centrifugal barrier displays a significant influence on the $|M| = 1$ states in a certain sense[26] and this term cannot be neglected. In our experiments, however, $\hat{F}^{1/4} = 1/80$ and the centrifugal term in the scaled Hamiltonian is two orders of magnitude smaller than the other terms. In this case, $l_z = 0$ is a good approximation and the centrifugal term can be omitted from the Hamiltonian.[17]

After neglecting the centrifugal term, the Hamiltonian above will not depend on the applied field or excitation energy separately. Instead, all the dependence is factorized into a single quantity, the scaled

energy $\varepsilon = EF^{-1/2}$. If we control the scaled energy ε to be constant by matching the exciting photon energy and the electric field at the same time, the classical dynamics of the atoms will be held invariant while, experimentally, we can record the absorption spectrum by scanning the excitation photon energy, which provides dynamical information about the outer electron. This procedure is successfully incorporated into the closed-orbit theory developed by Du and Delos.[1−3] According to this theory, the photon absorption rate is related to average oscillator-strength density $\overline{Df}(E,F)$, which can be written as a combination of a smooth background term and a set of sinusoidal oscillations,

$$\overline{Df}(E,F) = \overline{Df_0}(E) + \overline{Df_{\rm osc}}(E,F). \qquad (7)$$

The smooth background $\overline{Df_0}(E)$ is the basic oscillator strength in the absence of electric field, and the second term $\overline{Df_{\rm osc}}(E,F)$ is the contribution of the oscillations in the electric field. Defining $\omega = 2\pi F^{-1/4}$, the latter oscillatory part $\overline{Df_{\rm osc}}(E,F)$ can be written as

$$\begin{aligned}&\overline{Df_{\rm osc}}(E,F)\\ &= \sum_n (2\pi)^{1/2}\omega^{-1/2} D_0^n(\varepsilon)\sin[\hat{S}_0^n(\varepsilon)\,\omega - \Phi_0]\\ &\quad + \sum_{n,k} D_k^n(\varepsilon)\sin[\hat{S}_k^n(\varepsilon)\,\omega - \Phi_{nk}],\end{aligned} \qquad (8)$$

where $D_0^n(\varepsilon)$ and $D_k^n(\varepsilon)$ are the amplitudes of the nth repetition of the kth closed orbit for different initial conditions. In the calculations the maximum action is confined by the experimentally accessible precision $\Delta\omega$ according to the formula $S \le 2\pi/\Delta\omega$. In our experiment, the resolvable maximum action $S \le 12$, which means that the closed orbits with longer periods cannot be resolved under our conditions and a cutoff function is adopted to take the uncertainty in scaled energy into account.[25]

3. Experimental setup

The experimental setup has already been described in our previous papers[10,11] and only a brief introduction will be given here. An atomic beam produced from the resistively heated oven ejects into the interacting region, intersecting with the laser beam perpendicularly. A pinhole between the atomic oven and the interacting area serves to collimate the atomic beam. The interacting region is situated between two parallel Stark plates on which an appropriate electric voltage is applied. The laser beam comes from a Nd:YAG pumped dye laser system (Spectra Physics), frequency-doubled by a BBO crystal, and its wavelength is monitored by a Burleigh wavemeter. One photon is enough to excite the barium atom from the ground state $6s^2$ to the $6sn$p Rydberg state. The polarization of the laser beam is perpendicular to the direction of the electric field so that the $|M| = 1$ final state can be accessed. The strength of the electric field is computer controlled to keep the scaled energy constant. The excited atoms are ionized by electric field and the ions detected by a channel electron multiplier. The analog signal is recorded and accumulated in a boxcar and the final averaged digital data are downloaded into the computer for future processing.

4. Results and discussion

The experiment is done at three different scaled energies: $\varepsilon = -2.000$, -2.500 and -3.000. They cover an energy range of principal quantum numbers from $n = 33$ to $n = 43$ and the estimated uncertainty of the scaled energy is $\delta\varepsilon = 0.002$. The experimental spectrum is measured with respect to the wavelength of the photon at a fixed scaled energy and then undergoes a Fourier transform with respect to $F^{-1/4}$. The experimental and calculated recurrence spectra of barium at these scaled energies are shown in Figs.1, 2, 3 and 4, respectively. The uncertainty of the scaled energy is not considered in the spectral calculation based on the pure Coulomb potential of hydrogen, which shows all possible classical orbits.

Figure 1 shows the experimental and calculated recurrence spectra of barium at $\varepsilon = -2.500$. The experimental observation agrees well with the calculation based on the model potential for barium. The peaks (1)–(4) in Fig.1(a) are almost identical to the calculated result for barium on a model potential and for a hydrogen atom with pure Coulomb potential, as shown in Figs.1(b) and 1(c), respectively. Other peaks, prominently displayed in the calculated result for the hydrogen atom, are strongly suppressed as shown in Fig.1(a). When an uncertainty in scaled energy is taken into the barium closed orbit calculation, we find the recurrence intensity of orbits with large scaled action also weakens, as shown in Fig.1(b). This shows that the long time evolution orbits are very sensitive to experimental resolution. A coarse resolution conceals the contribution of long time evolution

orbits.

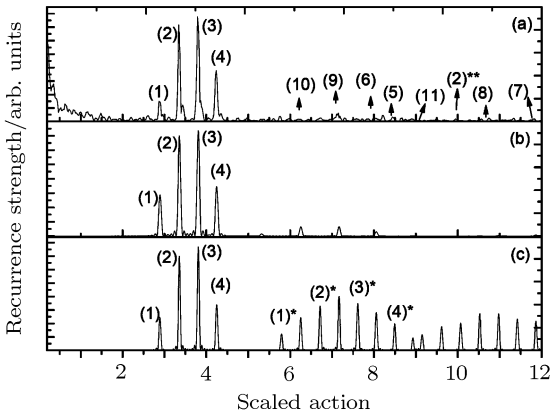


Fig.1. Experimental and calculated recurrence spectra of barium at $\varepsilon = -2.500$: (a) the experimental measurement, (b) the calculated spectrum with closed orbit theory based on the model potential for barium, and (c) the calculated spectrum with closed orbit theory for the hydrogen atom. The orbits corresponding to the marked peaks are displayed in Fig.2. The starred peaks correspond to the contribution of the repeated orbits.

The calculated closed orbits in $\rho - z$ coordinates with model potential are shown in Fig.2, corresponding to the labeled peaks in Fig.1. The orbits (1)–(4), as shown in Fig.2, all have short periods and make the main contribution for the stability of the corresponding recurrence spectral lines. This general feature of short period closed orbits has been seen in other experiments.[10,11] Additionally, the strength of the peak (1) in Fig.1 is slightly smaller than the calculated ones with model potential and with hydrogen pure Coulomb potential, which will be discussed later.

The orbits with large scaled actions are partly shown in Fig.2 and they are unstable for a long evolution time. Three types of orbits are worthy of special attention to understand the instabilities. (i) 'Snake' orbits. Snake orbits, for example, the orbits (9), (10) and (12) start out and return along the same trajectories and they are very sensitive to uncertainty in experimental conditions, even though some of them have simple moving patterns. They seem to have the largest magnifying factor for the perturbation from environment, and the corresponding recurrence intensities will be reduced accordingly. (ii) Long orbits. Long orbits accompanied by large actions form closed orbits after long time evolutions. The impact of external perturbation gradually accumulates and makes the orbit unstable. (iii) Repeated orbits. These kinds of orbits will penetrate the atom core at least once and are easily deviated by perturbations in experimental conditions. Their corresponding recurrence spectral intensities are strongly weakened by uncertainty in experiments as well. The repeated orbits are not shown in Fig.2 since they simply repeat their trajectories. Apparently, all orbits with longer periods usually correspond to larger scaled actions, are more sensitive to the environment and are unstable.

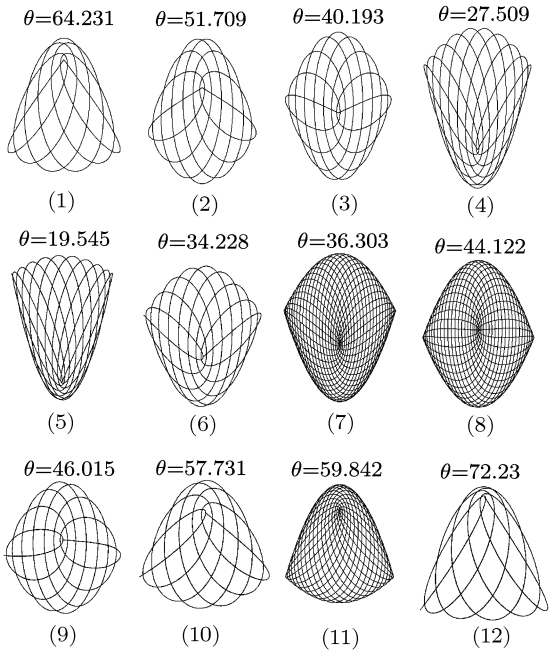


Fig.2. The calculated closed orbits in $\rho - z$ coordinates with model potential at $\varepsilon = -2.500$, corresponding to Fig.1. The orbits (1)–(4) are analogous to hydrogen ones and the others are suppressed strongly in recurrence intensities. The initial angles θ for the closed orbits are given in degrees.

In addition, we see some peaks with large actions yet these are missing in the spectrum calculated for the hydrogen atom. Even though the uncertainty of the scaled energy and the core effects are taken into account, they cannot be theoretically reproduced (Fig.1(b)). They may arise from combinatorial orbits due to inelastic core scattering events.[27,28]

The recurrence spectrum at lower scaled energy, $\varepsilon = -3.000$, is similar to the one at $\varepsilon = -2.500$, but the agreement with the calculated result is not as good, as shown in Fig.3. In the experimental spectrum as displayed in Fig.3(a), the peak (7) has a significant amplitude but it is missing in the calculated spectrum by model potential (Fig.3(b)). Another difference is that the peaks (1), (3) and (4) in the observation are much smaller than the calculated amplitudes based on either model potential (Fig.3(b)) or the hydrogenic

atom (Fig.3(c)).

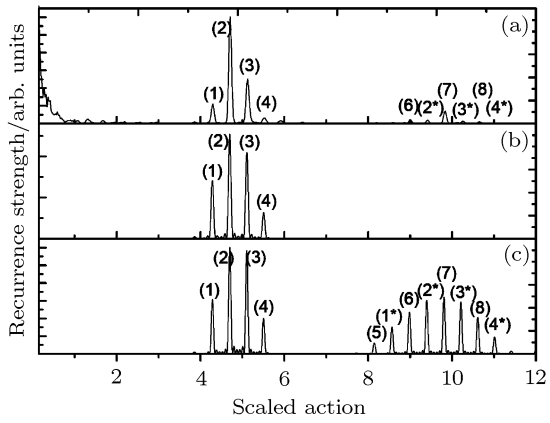


Fig.3. Experimental and calculated recurrence spectra of barium at $\varepsilon = -3.000$: (a) the experimental spectrum, (b) the spectrum calculated with closed orbit theory based on the model potential for barium, and (c) the spectrum calculated with closed orbit theory for the hydrogen atom.

Figure 4 is the recurrence spectrum of barium at higher scaled energy, $\varepsilon = -2.000$. As the energy approaches the classical limit, more orbits could be found. Here only the significant peaks are marked out. Similar to the observation at scaled energy $\varepsilon = -2.500$ (see Fig.1), there still remain some interesting peaks, such as the peak at $S = 3.75$ in Fig.4, which are missing in the theoretical calculations with hydrogenic potential. Compared to the spectrum at $\varepsilon = -2.500$, a peak with smaller scaled action, about 1.185, is found to have a stronger intensity than the calculated one by using either model potential or hydrogen potential. However, in contrast, the peaks (6), (7) and (9) show smaller intensities than the calculated one by using the model potential of barium. These differences cannot be explained even with the introduction of a scaled energy uncertainty that depresses the amplitude of the recurrence peak as at other scaled energies, i.e., $\varepsilon = -2.500$ and $\varepsilon = -3.000$.

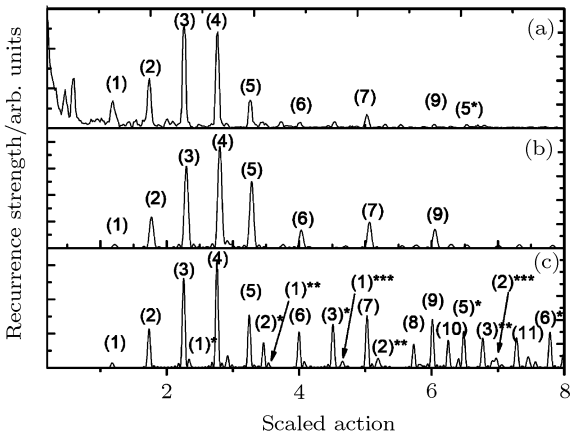


Fig.4. Experimental and calculated recurrence spectra of barium at scaled energy $\varepsilon = -2.000$: (a) the experimental spectrum, (b) the spectrum calculated with closed orbit theory based on the model potential for barium, and (c) the spectrum calculated with the closed orbit theory for the hydrogen atom.

This discrepancy can be explained from the relationship between quantum defect induced phase shift and the excitation energy, as shown in Fig.5. The 5d8p state of barium perturbs the 6snp state so strongly that the quantum defect induced phase shift abruptly changes at a wavelength around $\lambda = 238.45$ nm. It will cause, for example, at scaled energy $\varepsilon = -2.000$, the recurrence peaks with small scaled actions, i.e., peaks (1), (2) and those with larger scaled actions, i.e., peaks (7) to (9), to arise from different excitation energy ranges in which the perturbation of 5d8p state is different. This tendency can also be seen from the peak (3) which has a higher amplitude than the peak (4) in the measured spectrum (Fig.4(a)) while the intensities of the corresponding peaks are reversed in the calculated spectra based on model potential and hydrogen potential (Fig.4(b) and 4(c)).

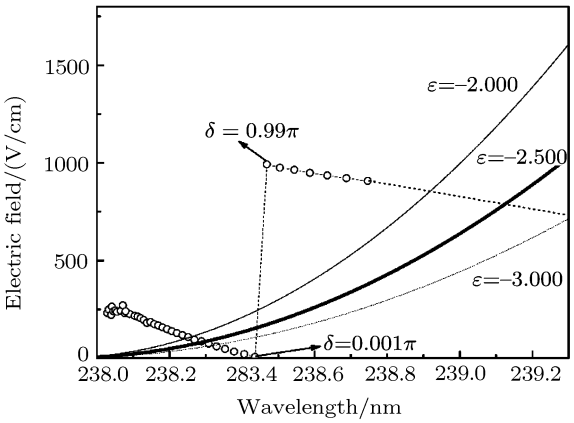


Fig.5. Energy range for the recurrence spectra of barium at scaled energy $\varepsilon = -2.000$, -2.500 and -3.000, and their relationship with the quantum defect μ. The 5d8p state of barium perturbs the 6snp state so strongly that the quantum defect induced phase shift abruptly changes at a wavelength around $\lambda = 238.45$ nm, as shown in an appropriate scale. The dotted line is extrapolated for the phase shift contained in the experimental data.

In comparison, however, the scaled energy spectrum of the strontium atom in an electric field does not differ so much from the calculated one for hydrogen as barium does. In the $|M| = 1$ configured excitation for strontium,[10] the scaled energy spectrum at $\varepsilon = -3.00$ is found to agree with the hydrogen spectrum, especially for the peaks around $S = 4.5$. Even

at larger scaled actions around $S = 9.5$, the peaks remain well reproduced by the calculations for the hydrogenic atom. In contrast, the first peak for barium at $\varepsilon = -3.000$ as in Fig.3(a) is suppressed by nearly one half and the peaks at higher scaled actions around $S = 9.5$ almost vanish. For the recurrence spectrum of strontium at $\varepsilon = -1.88$, unfortunately, it is not easy to arrive at an unambiguous conclusion due to the poor resolution of the spectrum. For $\varepsilon = -2.000$, as shown in Fig.4(a), the peak (1) at $S = 1.2$ is much larger than the calculated one while the peak (5) at $S = 3.3$ is suppressed by about one half. Moreover, the peak (3), instead of the peak (4) becomes the strongest one in the calculated spectra, which is different from the spectra of strontium at $\varepsilon = -3.00$ and $\varepsilon = -2.50$. The strong perturbation of 5d8p makes the energy-dependent quantum defects manifest more prominently in the scaled-energy spectrum of barium.

The failure of the model potential introduced in the Rydberg alkali atoms for the barium spectrum indicates that this model should be improved to fit the multi-valence atoms. As the alkali atom has only one valence electron, the model potential can well describe the quantum defects for different angular moment states.[7] For atoms with more valence electrons, however, this picture fails as the quantum defects are correlated with the main quantum numbers and are not constants any more.[8−11] In this sense, we hope the spectral observation for strontium and barium stimulate theoretical development to find its way to make better calculations. Experimentally, of course, we can combine the scaled-energy spectroscopy with the model potential for alkali atoms to reveal the effect of n-dependent quantum defects in atoms with complex electron configurations.

5. Conclusion

We have measured the scaled energy spectra in an electric field at $\varepsilon = -2.000$, -2.500 and -3.000 for barium. Closed orbit theory calculations are also carried out in an attempt to compare with the experimental recurrence spectra. The positions of the peaks are found to match well with each other. Some discrepancies remain for the amplitudes of spectral peaks, despite the use of the improved model potential for barium in our calculation. We believe the failure is due to the dependence of quantum defects on principle quantum number n, which consequently distorts the orbit of the electron and changes the corresponding recurrence spectrum. Conversely, however, we can experimentally explore the effect of n-dependent quantum defects in atoms with complex electron configurations in an external field.

Acknowledgments

We acknowledge Prof. Li You for helpful discussions.

References

[1] Du M L and Delos J B 1987 *Phys. Rev. Lett.* **58** 1731
[2] Du M L and Delos J B 1988 *Phys. Rev.* A **38** 1913
[3] Du M L and Delos J B 1988 *Phys. Rev.* A **38** 1896
[4] Gao J, Delos J B and Baruch M 1992 *Phys. Rev.* A **46** 1449
[5] Gao J and Delos J B 1992 *Phys. Rev.* A **46** 1455
[6] Gao J and Delos J B 1994 *Phys. Rev.* A **49** 869
[7] Weibert K, Main J and Wunner G 1998 *Ann. Phys.* **268** 172
[8] Kuik G J, Kips A, Vassen W and Hogervorst W 1996 *J. Phys.* B **29** 2159
[9] Kips A, Vassen W and Hogervorst W 2000 *J. Phys.* B **33** 109
[10] Zhan M S, Liu X J, Cao J W and Connerade J P 2001 *J. Phys.* B **34** 1175
[11] Liu X J, Cao J W, Zhan M S and Connerade J P 2002 *J. Phys.* B **35** 2069
[12] Gao J and Delos J B 1997 *Phys. Rev.* A **56** 356
[13] Connerade J P, Zhan M S, Rao J and Taylor K T 1999 *J. Phys.* B **32** 2351
[14] Eichmann U, Richter K, Wintgen D and Sandner W 1988 *Phys. Rev. Lett.* **61** 2438
[15] Courtney M, Jiao H, Spellmeyer N, Kleppner D, Gao J and Delos J B 1995 *Phys. Rev. Lett.* **74** 1538
[16] Wang W P, Li H Y, Wang S B and Lin S L 2002 *Chin. Phys. Lett.* **19** 29
[17] Kips A, Vassen W, Hogervorst W and Dando P A 1998 *Phys. Rev.* A **58** 3043
[18] Keeler M and Morgan T J 1998 *Phys. Rev. Lett.* **80** 5726
[19] Kips A, Vassen W and Hogervorst W 1999 *Phys. Rev.* A **59** 2948
[20] Keeler M L and Morgan T J 1999 *Phys. Rev.* A **59** 4559
[21] Wang D H and Lin S L 2004 *Chin. Phys.* **13** 464
[22] Bates K, Masae J, Vasilescu C and Schumacher D 2001 *Phys. Rev.* A **64** 033409
[23] Murray-Krezan J, Kelly J, Kutteruf M and Jones R 2007 *Phys. Rev.* A **75** 013401
[24] Connerade J P and Lane A M 1988 *Rep. Prog. Phys.* **51** 1439
[25] Main J, Wiebusch G, Welge K, Shaw J and Delos J B 1994 *Phys. Rev.* A **49** 847
[26] Wang W P, Li H Y, Wang S B and Lin S L 2008 *Chin. Phys.* B **17** 801
[27] Liu H P, Quan W, Shen L, Connerade J P and Zhan M S 2007 *Phys. Rev.* A **76** 013412
[28] Zhao L B and Du M L 2007 *Phys. Lett.* A **363** 453