Nonlinear spectroscopy of barium in parallel electric and magnetic fields*

Yang Hai-Feng(杨海峰)[a)b)], Gao Wei(高 伟)[b)], Cheng Hong(成 红)[b)], and Liu Hong-Ping(刘红平)[b)†]

[a)] College of Physics and Electronic Information, Luoyang Normal University, Luoyang 471022, China
[b)] 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 21 March 2014; revised manuscript received 11 April 2014; published online 10 August 2014)

We report on the experimental spectral observations of barium in parallel electric and magnetic fields. The laser pulse is linearly polarized along or perpendicular to the fields, leading to the states $m = 0$ and the states $m = \pm 1$ populated, respectively, by one photon excitation. By sweeping the electric field, we observe the linear and nonlinear splitting of the diamagnetic spectrum as the electric field increases. The spectral anticrossing is induced by the atomic core effect. The Stark spectrum also shows an obvious nonlinear quadratic behavior when the applied magnetic field varies strongly. All spectra are well explained by the full quantum calculation after taking the quantum defect effects of the channel ns up to nf into account.

Keywords: Rydberg atoms, Stark effect, diamagnetic effect

PACS: 32.30.–r, 32.60.+i, 03.65.Ge **DOI:** 10.1088/1674-1056/23/10/103201

1. Introduction

The Rydberg atom in an electric field or/and a magnetic field has been investigated for many years.[1,2] Due to the separability and hence integrability, hydrogen in an electric field is one of the least complicated systems. It helps us to understand the Stark effect at a higher level.[1,3,4] The non-hydrogenic atoms owning a finite sized ionic core, however, have many new features in an electric field, such as the level anticrossings in the Stark map.[5,6] For an atom in a strong magnetic field, the diamagnetic term in the Hamiltonian makes the system non-separable even for a hydrogen atom.[2] Researchers have developed a semiclassical method[7,8] and quantum calculation[9,10] to treat this problem and explain the correlative experiments.[11,12] Although there is a quadratic term in the Hamiltonian, the atom in a magnetic field has a simpler structure[13,14] than the Stark spectrum[6,15] for its higher symmetry.

The atom in combined electric and magnetic fields has also been studied by circularly polarized laser pulses.[16,17] Connerade *et al.* introduced a new experimental approach and analysis method.[16,17] In their experiments, they applied a σ^+ or σ^- polarized laser pulse along the magnetic field so that the circularly polarized components can be separated in the spectrum. With the electric field increasing, new spectral lines can break up from the "cluster of seed states",[16] similar to the hopf bifurcation due to chaos.[18]

Rather than using the circularly polarized laser pulse, in this paper, we linearly polarize the laser pulse along and perpendicular to the magnetic field. These spectra can provide complementary information on the interaction of an atom with the external fields. Unlike the research on the spectral structure of lithium in parallel electric and magnetic fields,[19,20] in our work, we concentrate on the evolution of diamagnetic spectra/Stark structures as the electric/magnetic field increases.

2. Experimental and theoretical treatment

In the experiment, we use the same apparatus described previously,[14] except for an additional electric field parallel to the magnetic field. An atom beam produced from an atom oven travels to the interacting point, intersecting with the laser beam perpendicularly in the center of the superconductor magnet (Oxford Instrument). Two pin-holes are inserted between the atomic oven and the interacting point to collimate the atomic beam. The interacting point is situated between a pair of electric Stark plates parallel to the magnetic field, where an additional electric field can be applied. One photon corresponding to 238 nm is used to excite the barium atom from the ground state to high Rydberg state 6snp and a pair of electric grids is used to ionize the excited Rydberg atom. The ionized residual is detected by a pair of microchannel plates mounted at the end of the chamber. The signal is recorded by an oscilloscope (Tektronix TDS1012) and the digital data stream is transferred to a personal computer for real-time signal processing. The laser beam is produced by a dye laser system (Lambda Physik, Scanmate 2E) pumped by Nd:YAG laser (Spectra Physics) and then frequency-doubled

*Project supported by the National Basic Research Program of China (Grant No. 2013CB922003) and the National Natural Science Foundation of China (Grant Nos. 11174329 and 91121005).

†Corresponding author. E-mail: liuhongping@wipm.ac.cn

© 2014 Chinese Physical Society and IOP Publishing Ltd

by a system-controlled beta barium borate crystal. A Soleil–Babinet compensator is used to rotate the laser from horizontal polarization to vertical polarization. Under this configuration, we can arrange the laser polarization parallel or perpendicular to the external fields to get π- or σ-polarized spectrum for barium in parallel fields.

Theoretically, the semi-classical treatment for a hydrogen atom has been developed to analyze the experimental spectrum.[19,21] For the problem of hydrogen in parallel fields, an approximate constant of motion $\Lambda_{\rm p}$ can be found for the weak field limit:[21]

$$\Lambda_{\rm p} = 4A^2 - 5A_z^2 + 10\beta A_z, \tag{1}$$

where $\boldsymbol{A} = \boldsymbol{L} \times \boldsymbol{p} + \boldsymbol{r}/r$ is the Runge–Lenz vector and $\beta = 12F/5B^2n^2$ a parameter to measure the relative strength of the electric and magnetic fields. Neglecting the paramagnetic interaction, for highly excited manifolds the energy shift can be written as[19]

$$\Delta E = B^2n^4(1 + \Lambda_{\rm p} + m^2/n^2)/16. \tag{2}$$

By quantizing $\Lambda_{\rm p}$ semiclassically, three different types of states can be classified to describe the contribution of the electric field on the diamagnetic spectrum of the atom.[22] The type-1 and type-2 states arise from the two-fold degenerate librational states in the lower part of the diamagnetic spectral line ($\beta = 0$) and give rise to a linear Stark effect as the electric field increases ($\beta \neq 0$). Moreover, the type-2 states will retrogress to the type-1 states at $\beta = 0.2$. The type-3 states arise from the non-degenerate rotational states in the upper part of the diamagnetic manifold and present a weak quadratic Stark effect as the electric field increases. All type-3 states will degrade into the type-1 states at $\beta = 1$.

However, to quantitatively describe the spectral structure, we have to take the quantum defect effects into account and include their contribution into the quantum calculation. For the Rydberg atom in parallel fields, we can define the z axis along the electric field or the magnetic field. The Hamiltonian for the outer electron of an alkali-earth metal atom, in atomic units, can be written as (in spherical polar coordinates)

$$H = \frac{p^2}{2} + V_r + \frac{B}{2}L_z + \frac{B^2}{8}\rho^2 + Fz, \tag{3}$$

where F, B are the electric and magnetic field strength, respectively, and V_r the Coulomb potential considering the polarization effects of the atomic core. The third and fourth terms are the paramagnetic and diamagnetic interactions, respectively. The last term is the interaction of the atom with the electric field. The core-induced electron screening effect in non-hydrogenic atoms makes the Schrödinger equation lose analytical solutions. In this case, the term V_r is usually expressed by model potential,[23] parameterized by some angular momentum-dependent variables, which are closely correlated with quantum defects. Rather than using the model potential V_r[23] or the R-matrix method including the quantum defects implicitly,[24] we employ an equivalent form by modifying the centrifugal potential[25]

$$V_r = \frac{\lambda(\lambda+1) - l(l+1)}{2r^2} - \frac{1}{r}, \tag{4}$$

where $\lambda = l - \delta + {\rm Int}(\delta)$ and the quantum defects are explicitly enclosed. Here ${\rm Int}(\delta)$ is the rounded nearest integer value of the quantum defect. In order to reproduce the experimental observation, a direct diagonalization of the Hamiltonian matrix is adopted. In the case of parallel electric and magnetic fields, the radial wavefunction is expanded in terms of B-spline basis while the angular wavefunction expanded in the normalized associated Legendre functions. It is described in detail in our previous work.[13] The magnetic quantum number is specified in advance since it is a good quantum number in our case.

In the calculation, the parameters of the effective quantum defects of different channels should be given as inputs.[10] In the examined energy range, they are determined as $\delta_{\rm s} = 0.29$, $\delta_{\rm p} = 0$, $\delta_{\rm d} = -0.34$, $\delta_{\rm f} = 0.04$, and $\delta_{\rm g\cdots} = 0$. For our excitation scheme of one-photon irradiation from the ground state $6{\rm s}^2\,{}^1{\rm S}_0$, only $6{\rm s}n{\rm p}\ {}^1{\rm P}_1$ is populated. It should be noted that the quantum defect values of barium are energy-dependent but luckily they do not change so quickly with energy. In this case, we can set their values fixed in the investigated energy range and the calculation gives reasonable and acceptable results.

3. Results and discussion

In order to study the effect of the parallel fields on the barium atom, we investigated the diamagnetic spectrum at various electric fields and the Stark spectrum at various magnetic fields as well. The ground state of barium is $6{\rm s}^2\,{}^1{\rm S}_0$. If the laser pulse is polarized along the electric and magnetic fields, the excited Rydberg states will have the same magnetic quantum number $m = 0$. While if the laser pulse is polarized perpendicular to the fields, both $m = 1$ and $m = -1$ states can be excited and the spectral line can be viewed as the combination of the pure $m = 1$ and $m = -1$ spectra.[14] The spectrum is recorded in the energy range including the manifolds of $n = 28$ and $n = 29$. The theoretical calculation of the spectrum lines have been convoluted with the Gaussian broadening of 0.15 cm^{-1} due to the laser linewidth and thermal atom Doppler broadening. The calculated spectra reproduce the experimental observations satisfyingly.

In Fig. 1 we plot the experimental diamagnetic spectrum at various electric fields. In the absence of an electric field, the magnetic field mixes only states with the odd parity since the transition occurs via one-step excitation from the ground state. The quantum defects of the $6{\rm s}n{\rm p}\ {}^1{\rm P}_1$ series are nearly integer

4 and that of the other odd parity channels are negligible, thus the diamagnetic spectrum at $F = 0$ shows more hydrogen-like features. When the electric field is on, the states with even parity are involved, resulting in the intrusion of the ns and nd states. For example, the state 31d emerges at $F = 20$ V/cm as shown in Fig. 1. When F increases, the state 31d interacts with the hydrogenic manifold of $n = 28$ and merges into it at $F = 60$ V/cm, leading to the redistribution of line intensity. It is different from the case of lithium where the quantum defect of channel d is negligible.[19] Examining the evolution of the lower states (the states in the lower energy side of the n-manifold), a linear Stark shift of the lower states profile is identified as F increases, which is plotted with a solid line. They correspond to the type-1 states defined in the semiclassical treatment.[22] While for the upper states (the states in the upper energy side of the n-manifold), a weak quadratic Stark effect as F increases is observed, which is plotted with dash lines. These states are defined as type-3 in the semiclassical method.[22]

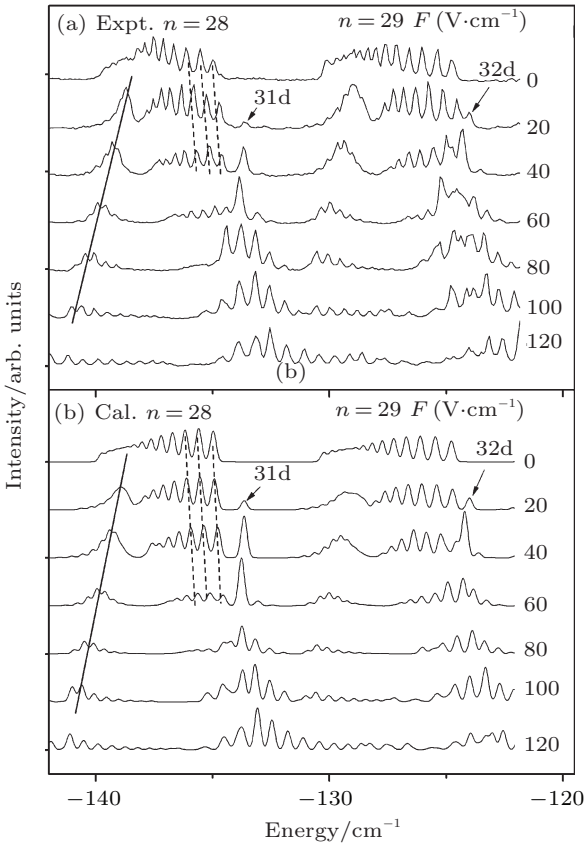


Fig. 1. Experimental (a) and theoretical (b) $m = 0$ spectrum of barium in the presence of parallel electric and magnetic fields. The magnetic field is kept at a constant value of 2.7 T while the electric field sweeps with a step of 20 V/cm. The calculated spectrum has been convoluted with a Gaussian broadening of 0.15 cm^{-1}. Evolutions of different states are plotted by a solid line and dash line.

To reveal the fine structure at lower electric fields, we calculate the spectrum of barium only for the manifold $n = 28$, $m = 0$. It is shown in Fig. 2, where the electric field F varies from 0 to 9 V/cm while B is fixed at 2.7 T. Under this condition ($\beta < 0.04$), all three types of states exist at the same time. The regions belonging to different types of states are separated by solid lines. Solid lines are determined by equations $\Lambda_p = -1 - 10\beta$, $\Lambda_p = -1 + 10\beta$, and $\Lambda_p = 25\beta^2$ respectively,[19] where $\beta = 12F/5B^2n^2$ and relation between energy shift and Λ_p is determined by Eq. (2). The states of different types display completely different behaviors as F increases. Obviously, in region III, the states of type-3 are scarcely perturbed by the electric field and present the nonlinear shifts when F increases. While in region I, the states of type-1 display a linear Stark shift towards the low energy side. This can be interpreted semiclassically. The lower states in the diamagnetic manifold are two-fold degenerate and librationally symmetric. Their wavefunctions are strongly stretched along the field direction and very sensitive to the applied electric field.[19] In the mediate region II, the states of types 1 and 2 appear simultaneously, behaving rather irregularly. For example, new spectral components give birth and the line intensity distribution varies rapidly with F. Another notable feature is the present of level anticrossing in the mediate region II. Tracing the states labeled by arrows in Fig. 2 as F increases, it can be seen that the oscillator strength decreases until $F = 4$ V/cm, and increases again. Conversely, the states on the other side of the dash line present an opposite process that the oscillator strength increases first and then decreases. This is the vivid manifestation of the level anticrossings induced by the ionic core in barium. The linear or nonlinear splittings of the diamagnetic spectrum can be expounded by the interaction between the states coupled by external fields. The magnetic field mixes the states with odd parities while the electric field couples the adjacent states with opposite parities. It is natural that the states come to receive degeneracy lifting and give birth to more spectral lines when the electric field is applied.

When the laser pulse is linearly polarized perpendicular to the electric and magnetic fields, both $m = 1$ and $m = -1$ states can be populated simultaneously. In order to study the magnetic field effect on Stark structures, we obtained a series of Stark spectra of $F = 56$ V/cm at different magnetic fields. The experimental and calculated spectra are shown in Fig. 3. For the σ-polarized spectrum ($|m| = 1$), the contribution from the s channel is absent compared with the π-polarized one. When the magnetic field is null, the states are two-fold degenerated. The non-zero magnetic field will lift the two-fold degeneracy, and the Stark spectra will split into a pair of lines corresponding to $m = 1$ and $m = -1$, respectively. The paired lines at $B = 1$ T are labeled by arrows in Fig. 3 and the intervals between them are about 0.93 cm^{-1}, equal to the energy gap between the positive and negative paramagnetic terms in the Hamiltonian. Examining the evolution of the Stark spectral lines, plotted by a dash line in Fig. 3, the non-hydrogenic state 32d presents a quadratic Zeeman effect due to the diamagnetic interaction. When the magnetic field increases further, since

the magnetic field can couple states with the same parity, the manifolds np and $(n-1)$d mix.

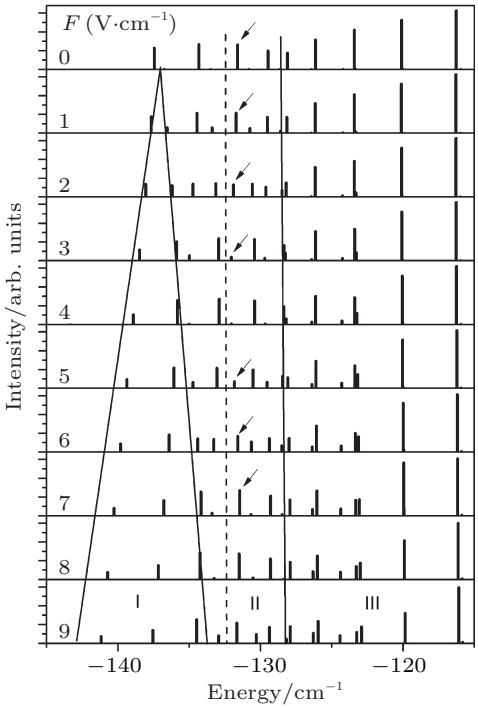


Fig. 2. Calculated bar spectrum of barium for the manifold n = 28, m = 0. The magnetic field is kept at the value of 2.7 T while the electric field sweeps with a tiny step of 1 V/cm. The spectral broadening is not included for uncovering the fine structure. The regions belonging to different types of states are separated by the solid lines. Level anticrossing between the states of types 1 and 2 occurs along the dashed line. See text for detail.

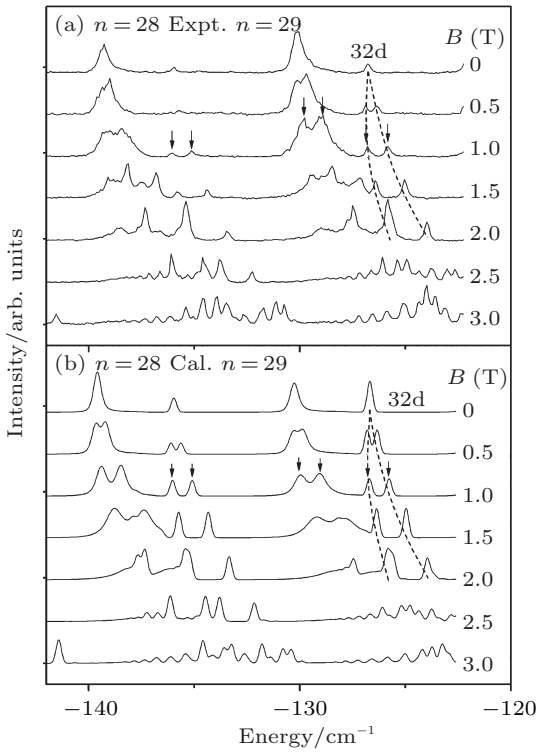


Fig. 3. Experimental (a) and theoretical (b) $m = \pm 1$ spectrums of barium in the presence of parallel electric and magnetic fields. The electric field is kept at 56 V/cm while the magnetic field sweeps with a step 0.5 T.

4. Conclusion

The spectral structure of a barium atom irradiated via one photon excitation in the parallel electric and magnetic fields is systematically investigated. The experimental setup allows the observation of the π and σ spectra by linearly aligning the laser pulse along or perpendicular to the fields. The full quantum calculation is also performed to explain the obtained spectra. The evolutions of π-polarized diamagnetic spectra are studied at various electric field strengths and three classes of states defined in semi-classical treatment are identified. The core effect of barium is carefully analyzed in the calculated spectra and the level anticrossing is identified unambiguously. As the electric field increases, the structures of the diamagnetic manifold present linear and nonlinear splittings. It is readily understood that the electric field mixes more states with opposite parity. The evolutions of the σ-polarized Stark structure as the magnetic field increases also show a nonlinear spectral splitting due to the removing of the degeneracy of $|m|$ by applying the magnetic field.

References

[1] Rojansky V 1929 *Phys. Rev.* **33** 1
[2] Zimmerman M L, Kash M M and Kleppner D 1980 *Phys. Rev. Lett.* **45** 1092
[3] Rice M H and Good J R H 1962 *J. Opt. Soc. Am.* **52** 239
[4] Richter K, Wintgen D and Briggs J S 1987 *J. Phys. B: At. Mol. Opt. Phys.* **20** L627
[5] Littman M G, Zimmerman M L, Ducas T W, Freeman R R and Kleppner D 1976 *Phys. Rev. Lett.* **36** 788
[6] Yang H F, Gao W, Cheng H and Liu H P 2013 *Phys. Scr.* **87** 035301
[7] Braun P A 1983 *J. Phys. B: At. Mol. Opt. Phys.* **16** 4323
[8] Delos J B, Knudson S K and Noid D W 1983 *Phys. Rev. A* **28** 7
[9] O'Mahony P F and Taylor K T 1986 *Phys. Rev. Lett.* **57** 2931
[10] Yang H F, Gao W, Quan W, Liu X J and Liu H P 2012 *Phys. Rev. A* **85** 032508
[11] Connerade J P, Droungas G, Elliott R, He X, Karapanagioti N, Farooq M A, Ma H, Marangos J P and Nawaz M 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 2753
[12] Elliott R J, Droungas G, Connerade J P, He X H and Taylor K T 1996 *J. Phys. B: At. Mol. Opt. Phys.* **29** 3341
[13] Gao W, Yang H F, Cheng H, Liu X J and Liu H P 2012 *Phys. Rev. A* **86** 012517
[14] Yang H F, Gao W, Cheng H and Liu H P 2013 *Chin. Phys. B* **22** 053201
[15] Dong H J, Wang T, Li C Y, Zhao J M and Zhang L J 2013 *Chin. Phys. B* **22** 073201
[16] Connerade J P, Droungas G, Karapanagioti N E and Zhan M S 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** 2047
[17] Connerade J P, Hogan S D and Abdulla A M 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** S141
[18] Ganesan K and Gebarowski R 1997 *Pramana J. Phys.* **48** 379
[19] Cacciani P, Liberman S, Luc-Koenig E, Pinard J and Thomas C 1988 *J. Phys. B: At. Mol. Opt. Phys.* **21** 3523
[20] Taylor K T, Nayfeh M H and Clark C W 1988 *Atomic Spectra and Collisions in External Fields* (Berlin: Springer)
[21] Cacciani P, Luc-Koenig E, Pinard J, Thomas C and Liberman S 1986 *Phys. Rev. Lett.* **56** 1467
[22] Cacciani P, Luc-Koenig E, Pinard J, Thomas C and Liberman S 1988 *J. Phys. B: At. Mol. Opt. Phys.* **21** 3499
[23] Wang L, Quan W, Shen L, Yang H F, Shi T Y, Liu X J, Liu H P and Zhan M S 2009 *Chin. Phys. B* **18** 4791
[24] Seipp I and Taylor K T 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 2785
[25] Simons G 1974 *J. Chem. Phys.* **60** 645