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Experimental investigation of barium spectra in crossed electric and magnetic fields*

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This paper has studied the barium spectra in external magnetic and electric fields on the recently-built system based on a super-conducting solenoid with high stability of better than 1 Gauss. Firstly, we compared the spectra in a high pure magnetic field of $B = 4.00000$ T for different polarized laser, σ^+ and σ^-. We prove that after a proper shift, the σ^+ and σ^- spectra are completely identical. We also investigated the dependence of the spectral feature in the high magnetic field on an additionally applied crossing electric field. Along with the increase of the electric field, there is an energy shift for every spectral line. Based on a transformative Hamiltonian, we explained this energy shift well.

Keywords: crossed fields, Rydberg, barium
PACC: 3150, 3345B, 3320L

1. Introduction

The high-lying, single-electron, Rydberg state of atoms in external static fields provides a low-dimensional prototypical physical system to study the basic problems of interest.[1−9] A typical example is the spectrum in a pure magnetic field or in crossed electric and magnetic fields, which is very intriguing since it is closely concerned with the quantization of non-integrable system based on the Gutzwiller trace formula.[10] In a pure magnetic field, the system has a rotational symmetry about the direction of the magnetic field and this rotational symmetry corresponds to a conservative physical variable, the orbital angular momentum. We can separate this orbital angular motion perpendicular to the magnetic field easily and reduce the problem to a two-dimensional one. For a Rydberg atom excited high enough, if the external magnetic field gets stronger and stronger, its influence will be comparable with the Coulomb interaction. At this moment, the system gets highly non-integrable and behaves classically chaotic. Moreover, in crossed fields, with the introduction of the electric field, the rotational symmetry will get lost and the Hamiltonian becomes non-separable at all, which makes the theoretical process more complicated. A semi-classical theory, closed orbit theory (COT), is firstly developed by Du and Gao to explain the absorption spectrum near the ionization threshold successfully.[5,11−14] This theory is also extended to take quantum defect into account.[15] In the atomic system with a small core, the Hamiltonian can be described as identical to the Hydrogen atom since the Rydberg electron is far away from the core and no collision occurs most of time. However, when the core gets bigger and bigger, the electron will be scattered with the core and the system cannot be seen as a hydrogen atom model simply,[15−19] especially for the large core atoms, such as barium we are interested in.

Owning to a low ionization energy ($IP = 5.212$ eV) and a two-free-electron open shell, barium spectrum in external fields has been investigated extensively since the brilliant work in a strong magnetic field performed by Garton and Tomkins at 1969.[20] In 1977, Fonck *et al* observed the diamagnetically induced states mixing between the 6sns $^1S_0$ and 6snd $^1D_2$ series and their energy shifts compared with the hydrogenic values.[21] The influence of high-lying doubly excited 5d8p perturber below the first ionization limit of barium on the level energies of the odd-parity

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Rydberg 6snp and 6snf levels was also studied.[22] The barium spectrum in a magnetic field of 2.89 T revealed an extra structure between the σ^+ and σ^- and it was proposed to be explained by the many-electron nature of the barium atom.[23,24] Rao and Taylor have also done a theoretical calculation for barium and hydrogen to identify the non-hydrogenic behaviour of barium.[25] However, the application of the refined closed-orbit theory, even taking the core effect of barium atom into account, can not explain the observed experimental spectrum well,[26] which remains a stimulating challenge for continuing this work. Afterwards, the spectrum of barium in an electric field was observed in a region where singly excited Rydberg states are heavily perturbed by a doubly excited state.[27] Using a high resolution, pulsed laser system and a crossing configuration for fields, other than the parallel one adopted by König,[28] we investigated the barium spectra at increasing electric field strength for a fixed magnetic field strength of 1 T by Connerade group[29] and observed an obvious change for the spectral profile with the increasing electric field. One year later, in the same group, they obtained barium spectra in crossed fields at shorter wavelength from $\lambda = 238.14$ nm to $\lambda = 238.30$ nm at lower magnetic field strength $B = 0.57$ T. The analysis of the nearest-neighbour level spacing statistics showed that the electron dynamics evolutes as the electric field strength is increased.[30] It seems that a higher magnetic field will make it available to reveal the state mixing in another point of view.

Using the recently-built laser–atom interaction system and a new super-conducting solenoid with high stability, we investigated the barium spectra in an increasing electric field for a fixed magnetic field of 4.00000 T, much higher than that reported before.[29,30]

2. Experimental

The experimental setup has already been described in our previous papers,[31] only a brief introduction will be given here. An atomic beam produced from a resistance-heated oven travels to the interacting point, intersecting with the laser beam at an angle of 15°. 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 crossed to the magnetic field, where an additional electric field can be applied. One photon is used to excite the barium atom from the ground state to high Rydberg state and a pair of electric grids is used to ionize the excited Rydberg atom. The ionized residual is detected by a pair of micro channel plates mounted at the end of the chamber. The signal registered by the oscilloscope will be downloaded to a computer for further process. The laser beam is produced by a dye laser system (Lambda Physik, Scanmate 2E) pumped by Nd:YAG (Spectra Physics) laser and subsequently frequency-doubled by a Barium Metaborate (BBO) Crystal. The wavelength is monitored by a Burleigh wavemeter and its scanning is controlled by the same computer. The super-conducting solenoid (Oxford) we used can rise to a magnetic field up to 6 T and sweep with stability of better than 1×10^{-4} T.

3. Results and discussion

Firstly we acquired the barium spectra in a static pure magnetic field with σ^+ and σ^- polarized laser, respectively. Compared with the previous works,[29,30] we performed our measurements at a higher magnetic field, $B = 4.00000$ T, since our laser has a broader bandwidth of 1.4 GHz. The spectra were recorded with much lower n values from $n = 33$ to $n = 43$. The obtained spectra in fixed magnetic field are presented in Fig.1. To compare the different polarized spectra, we have performed a proper paramagnetic shift for σ^- branch with $\Delta\lambda = 0.0215$ nm. As is well known, the shift is equal to the strength of the magnetic field in atomic unit if the spectra are obtained in a pure magnetic field. The Hamiltonian (in atomic units) of a Rydberg hydrogen atom in crossed electric and magnetic fields is:

$$H = \frac{1}{2}p^2 - \frac{1}{r} + \frac{1}{2}\gamma L_z - Fx + \frac{1}{8}\gamma^2(x^2 + y^2), \quad (1)$$

where $p^2 = p_x^2 + p_y^2 + p_z^2$, $r = \sqrt{x^2 + y^2 + z^2}$, γ is defined as $\gamma = B/c = B/(2.35 \times 10^5)$ with B in T. According to Eq.(1), after the cancellation of motional Stark effect although it is very small for our apparatus configuration, the shifted energy ΔE should be equal to $(1/2) \cdot \gamma \cdot |\Delta m| = (1/2) \cdot \gamma \cdot 2 = \gamma = 0.0213$ nm, in coincidence with the shifted value 0.0215 nm in our data process in Fig.1. As we can see from Fig.1, the spectra for σ^+ and σ^- after shifting are completely identical for the spectral line positions. It indicates that there is no perturbation in this excitation energy

range corresponding to wavelengths from 238.5500 nm to 238.1000 nm or that there is a perturbation but it has the same effect on energy shifting for both polarizations. Usually the spectral line intensity gets stronger and stronger with the increase of wavelength, but our observation is on the other way round. It indicates that there should exist a perturber, which proves to be 5d8p according to the earlier observation.[22] Even so, the two-electron correlation is not important in the photon energy range we scanned since the energy shift is identical for each spectral line, which will be discussed in the following parts.

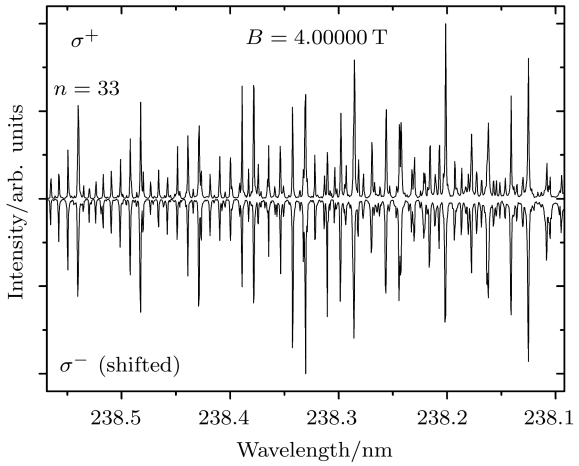


Fig.1. The comparison of σ^+ and σ^- spectra of barium in a constant magnetic field of 4.00000 T. After a proper shift of $\Delta\lambda = 0.0215$ nm, the two spectra are almost identical.

Another obvious phenomenon is that the spectral lines below the energy of photon $\lambda = 238.4500$ nm in free field are now split into sub-peaks which are labelled by different orbital angular momentum quantum number l although the main quantum number n is kept as a good one. As the increase of excitation energy, i.e., beyond the photon energy of $\lambda = 238.4000$ nm, the spectral lines corresponding to different quantum number l of one main quantum number n begin to overlap with those peaks of the neighbouring main quantum number $n'(n' \neq n)$. Specially, around wavelength $\lambda = 238.2000$ nm, the state mixture leads to the spectral intensity distribution into disorder, which means that the global symmetry is completely broken by the applied strong magnetic field. Based on the quantization scheme for the crossed-field problem established by Pauli, adopted by Connerade to explain their experimental result,[24] we find that the magnetic field induced energy shift, to first order, should be $\Delta E_{nk}(B) = (1/2) \cdot kB$, where k is the Pauli quantum number. So, for the neighbouring split lines of the same main quantum number n, the difference of the energy shifts is $\delta E_n = |(1/2) \cdot kB - (1/2) \cdot k'B| = (1/2) \cdot |\Delta k|B$. For our experimental condition of the high magnetic field, $B = 4.00000$ T, the value of $(1/2) \cdot B$ is calculated to be 0.0110 nm. On the other hand, the measured spectral lines spacing is around 0.0120 nm. These two values are very close, which indicates that the neighbouring Pauli quantum numbers should be $k, k \pm 1$.

As an additional applied crossed electric field, the rotation symmetry is destroyed and every manifold evolves along with the strength of the electric field. We use σ^- circularly polarized laser to excite the atom in crossed fields to investigate the effect of crossed external electric field on the spectral feature in high magnetic field of $B = 4.00000$ T as depicted in Fig.2. Since the magnetic field intensity is very high, we choose the spectra from $n = 33$ to $n = 37$ to study. The electric field is swept from zero to 140.00 $\mathrm{Vcm^{-1}}$ with step of 20.00 $\mathrm{Vcm^{-1}}$. At zero electric field, we can see that each spectral line in free field is split into several small peaks corresponding to $k = 0, 1, 2, ...$ due to the diamagnetic interaction as discussed before and the spectra are very easy to assign. When the electric field increases, typically up to $F = 40.00\,\mathrm{Vcm^{-1}}$, this spectra feature still keeps well. However, when the electric field exceeds $F = 60.00\,\mathrm{Vcm^{-1}}$, we can not pick out the peak $k = 0$ for a certain quantum number n any more. The k-mixing comes into effect and it is not easy to characterize the spectra according to the main quantum number n.[21]

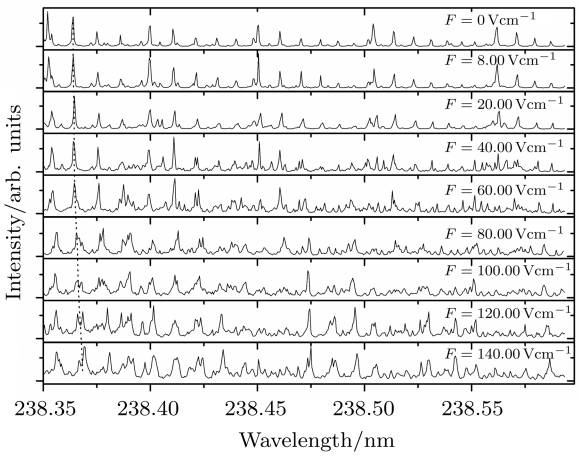


Fig.2. The effect of the externally applied electric field on the spectral feature in constant high magnetic field of 4.00000 T is demonstrated in dot line. The electric field is varied from zero to 140.00 $\mathrm{Vcm^{-1}}$.

A more interesting phenomenon is that when the electric field goes beyond $F = 100.00\,\mathrm{Vcm^{-1}}$, $k = 0$ peaks do not prevail any more and some strong peaks come up instead. When the electric field increases further, i.e., up to $F = 140.00\,\mathrm{Vcm^{-1}}$, this phenomenon extends to the whole spectral range. The dependence of the spectra in magnetic field of high intensity on the electric field is depicted in Fig.2 by dotted line for demonstration. According to the term γL_z in the Hamiltonian given in Eq.(1), we find that the first spectral splitting occurs for $m = 0, -2$ since our laser is σ^- polarized. It shows that the first state mixing begins from the neighbour components $m = 0, -2$ of the upper state $m = -1$. This is proved by quantum calculation, too.[22]

We can also rewrite the potential part V in Hamiltonian displayed in Eq.(1) as

$$V = -\frac{1}{r} + \frac{1}{2}\gamma L_z + \frac{1}{8}\gamma^2\left(x - 4\frac{F}{\gamma^2}\right)^2 + \frac{1}{8}\gamma^2 y^2 - 2\frac{F^2}{\gamma^2}, \quad (2)$$

and it can be seen that if the excited state is so high that its orbit is far away from the Coulomb centre ($x \gg 4 \cdot F/\gamma^2 \sim 250a_0$), the external electric field, combined with the magnetic field, will contribute additional energy, which results in an energy shift. As the first approximation, the energy shift is proportional to $2 \cdot F^2/\gamma^2$, which is drawn in Fig.3 in solid line. The parameter of magnetic field is taken as $B = 4.00000\,\mathrm{T}$ for our experimental condition. Taken out from Fig.2, the data points for the energy shift are also presented for comparison. We can see that the experimental energy shift has the same dependence on the applied electric field with the Eq.(2), but there is still some discrepancy as the electric field increases, especially up to $120.00\,\mathrm{Vcm^{-1}}$, which may be due to the fact that the excited energy is not high enough and this simple relationship will not come into existence accurately any more.

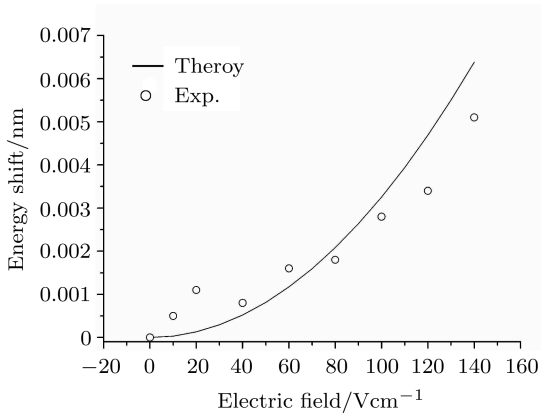


Fig.3. The dependence of the spectral line shift on the externally applied crossing electric field. The magnetic field is kept to a constant value of $B = 4.00000\,\mathrm{T}$. The solid line is drawn based on the transformative potential part $2 \cdot F^2/\gamma^2$ given in Eq.(2) and the data points are taken from our experimental results presented in Fig.2.

4. Conclusion

As the first experiment performed on our recently-built system to investigate some basic physical problems, the barium spectra have been studied in the high magnetic field of $B = 4.00000\,\mathrm{T}$. The σ^+ and σ^- spectra of barium in a constant magnetic field of $4.00000\,\mathrm{T}$, after a proper shift, prove to be almost identical. The dependence of spectral feature on the externally applied crossed electric field varying from zero to $140.00\,\mathrm{Vcm^{-1}}$ has also been investigated and the spectral line shift with the increasing electric field is well explained based on the transformative Hamiltonian.

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