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THE POTENTIAL MODEL INVESTIGATION OF STARK EFFECT IN CAESIUM RYDBERG STATE ATOMS AND COMPARISON WITH EXPERIMENT*

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The potential model method for computation of Stark structure of Cs Rydberg states atoms and oscillator strength is described, for external electric fields varying from 0 to 600V/cm. Anticrossing, l-mixing and n-mixing phenomena are observed clearly from the map of Stark. Corresponding experiment is performed under the same condition, and the two results are in good agreement with each other within the experimental uncertainty.

Keywords: PM method, Cs Rydberg states atoms, Stark structure
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I. INTRODUCTION

The study of Rydberg state atoms in external fields is always an important aspect of the atomic physics.[1,2] In Rydberg atoms, one of the outer electrons is highly excited and situated far from the atomic core. As is known, they have a lot of properties different from those atoms in the ground state, because of the large principal quantum number n. The binding energy of the core to Rydberg electron rapidly decreases, roughly as n^{-2}, while the coupling of the electron with the applied electric field has a relation of fn^2. Thus the influence of a weak electric field on the Rydberg electron can be equivalent to the Coulomb interaction, and the Stark effect can be significant to the behavior of highly excited atoms.

With the development of laser technology and calculation methods, experimental investigations and theoretical predictions on Rydberg atoms have been carried out extensively in recent years[3,4]. Littman *et al.*[5] studied lithium Rydberg atoms and found features different from hydrogen atoms. Zimmerman *et al.*[6] studied pure Stark effect of Rydberg alkali metal atoms at n=15. To our knowledge, no efforts have been made to study the Stark effect of heavier alkali metal atom such as caesium at higher $n > 15$. Meanwhile caesium is more attractive because of its heavy atomic core and being most nonhydrogen-like. In this paper we present our calculation results of the Stark effect and oscillator strength of caesium Rydberg atoms at n=21–23 in electric fields in a range of 0–600 V/cm by using the potential model(PM) method, and for comparison, we performed experiment under the same conditions.

II. POTENTIAL MODEL BASIS METHOD

A valence electron of Rydberg caesium in external electric fields can be described by the Hamiltonian

$$\mathcal{H} = -\frac{1}{2}\nabla^2 + V(r) + Fz. \tag{1}$$

We still use the conventional expansion method of basis set and diagonalizing the Hamiltonian matrix, but the PM basis set is employed in this case as in previous work.[7,8] In PM, the core potential of caesium atom takes the following analytic form

$$V(r) = -Z_p(r)/r = -\frac{1}{r}\left(1 + \frac{B'}{(r+\gamma')^2} + \frac{\kappa'}{(r+\xi')^2}\right), \tag{2}$$

and the accompanying radial wave function reads

$$R_{nl}(r) = N\exp(-\rho/2) \cdot \rho^s(\rho+\gamma)^l(\rho+\xi)^u \sum_{\mu=0} a_\mu \rho^\mu, a_0 = 1, \tag{3}$$

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where the potential parameters $\beta', \gamma', \xi', \kappa'$, wave function parameters s, t, u and expansion coefficients a_μ, all depending on n and l, are to be determined. Here $\rho = \alpha r, N = \alpha\gamma', \xi = \alpha\xi', a = (1+\delta_{nl})/n^*$, $n^* = n - \delta_{nl}$, and the quantum defect δ_{nl} is the sole input parameter which can be got from previous experimental data. Putting $V(r)$ and $R_{nl}(r)$ into the free-field radial Schrödinger equation

$$\left[-\frac{1}{r^2}\frac{\mathrm{d}}{\mathrm{d}r}\left(r^2\frac{\mathrm{d}}{\mathrm{d}r}\right) + \frac{l(l+1)}{r^2} + V(r)\right] R_{nl}(r) = E_{nl}R_{nl}(r), \tag{4}$$

we have to solve the resultant algebraic equations of all the above parameters, then get zero-field $R_{nl}(r)$ in its analytical form. So the potentials are nl-dependent and the total wave functions can be written as $\Psi_{nlm} = R_{nl}(r)Y_{lm}(\theta,\varphi)$, corresponding to PM. $Y_{lm}(\theta,\varphi)$ is the well-known spherical harmonical functions. This group of wave functions forms the complete basis set for PM method.

Expand the field-dressed wave function in terms of such PM zero-field eigenstates (PM basis) and diagonalize the Hamiltonian matrix to obtain the energies and the wave functions. The formula of energy matrix element is

$$H_{nlmn'l'm} = -\delta_{nn'}\delta_{ll'}/2(n-\delta_{nl})^2 + f < r\cos\theta >_{nlmn'l'm}, \tag{5}$$

where $\delta_{nn'}$ and $\delta_{ll'}$ are δ functions, atomic unit is adopted in external electric field $f = F/F_0$, $F_0 = 5.14\times10^9$ V/cm and F is the true value used in the experiment. The oscillator strengths for transitions from the ground state $|E_g lm\rangle$ to Stark manifolds $|E_k m\rangle$ are also worked out as

$$f_{E_g,E_k} = \frac{2}{3}(E_g - E_k)\left|\sum_{E'_k}\sum_{l'} U^{E_k}_{E'_k l'} \times \left\langle E_g lm|\boldsymbol{\varepsilon}\cdot\boldsymbol{r}|E'_k l'm\right\rangle\right|^2. \tag{6}$$

For our calculation on the Stark structure of Cs Rydberg atoms with the effective quantum number n^*=21–23 , the basis set is formed by selecting the PM wave functions in the range of $15 \le n \le 35$, and the size of energy matrix is 525×525.

III. EXPERIMENTAL SETUP

The experimental setup (see Fig.1) is composed of three parts: atomic beam, interaction region and signal detecting and handling. The system pressure is kept at the level of 10^{-5} Pa pumped by diffusion/mechanical pump pairs. Two dye lasers are used to create the Cs Rydberg states by two-step excitation (λ_1 = 852.1 nm, λ_2 about 515 nm pumped by harmonics of the same Nd:YAG laser). Ionization signals are detected by the channel electron multiplier (CEM) and stored or handled in a PC.

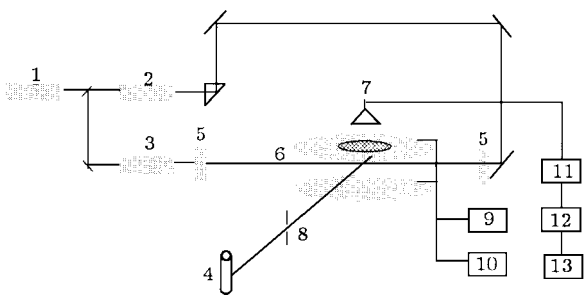


Fig.1. Schematic diagram of experimental apparatus 1—Nd:YAG laser, 2—Dye laser1, 3—Dye laser2, 4—Atomic oven, 5—Polarizer plate, 6—Electrode plate, 7—Channel electron multiplier, 8—collimationaperture, 9—DC power supply, 10—Pulse high-voltage supply, 11—Pre-amplifier, 12—Boxcar, 13—Personal computer.

IV. RESULTS AND DISCUSSION

In this section we present our calculations of the Stark structure and our experimental results of Cs Rydberg atoms. The results are displayed as a set of maps. We have worked in a single range of energy: $-340 - -240\mathrm{cm}^{-1}$ (defined as from 1st ionization limit) and electric field $0-600$ V/cm. Δm=0 was assumed. These values contain the n^*=21–23 manifold in fields from l-mixing to n-mixing region.

All energies were calculated for the embedded energy levels and the plots were generated in connection with the energy values corresponding to each selected field value. Figure.2 presents all the results, which illustrates the fan structure of Stark manifold corresponding to the change of applied external fields as well as the features of levels anti-crossing due to the heavier core effect. It also shows the trend of transition from l-mixing in low field strength to n-mixing in high field strength. At the same time, we can observe the levels expansion occurring first in the area of ^{2}S states, and an explanation can be got from the states with low l value possessing large quantum defects.

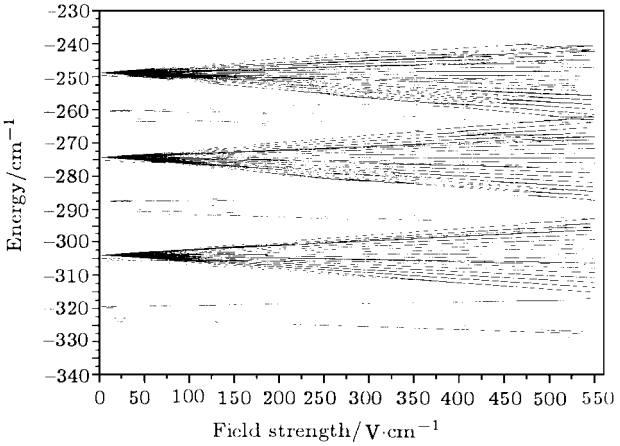


Fig.2. Caculated Stark structure of Cs ($n^* = 21-23$) with PM method.

Figure 3 includes eight typical field ionization spectra of Cs Rydberg atoms in the energy range of $n^* = 21-23$, the 2nd laser being scanned from 512nm to 519nm, and the high voltage being above the field ionization threshold. For convenience of comparison, we arrange the spectra in fields of 0, 170, 220, 270, 320, 370, 470, and 520V/cm together in the same figure. It clearly illustrates that in zero field, the energy line is separated and the signal intensity difference between S and D states shows that the transition possibility from 6^2P to D is lager than that to S. As the applied field increases, the transition from zero-field to l-mixing to even n-mixing can be clearly seen from the figure.

The agreement between the calculated and experimental results can be seen from Table 1 and Fig.4. One of the experimental results is listed in Table 1 with corresponding calculated values under the condition of 270 V/cm. As can be seen, fairly good agreement between them is obtained within ± 0.3cm^{-1}, which we can consider to be the experimental uncertainty in this study.

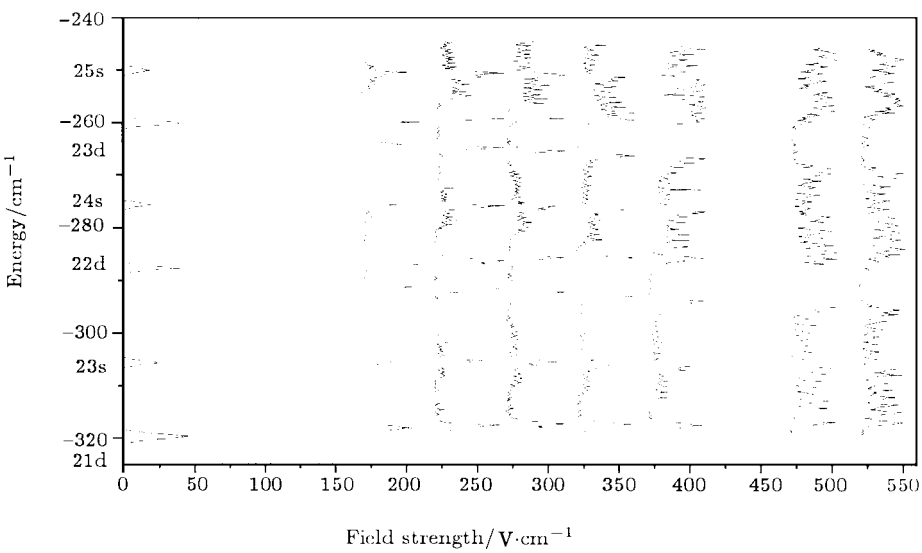


Fig.3. Experimental Stark spectra of Cs Rydberg atoms.

Table 1. Stark energy levels of caesium atoms with $n^* = 21-23$ in a external electric field 270 V/cm (unit: cm^{-1})

Theo. Value	Exp. Value	Errors	Theo. Value	Exp. Value	Errors	Theo. Value	Exp. Value	Errors
−317.8	−317.9	0.1	−285.6	−285.8	0.2	−265.4	−265.6	0.2
−309.2	−309.3	0.1	−279.9	−279.8	−0.1	−259.2	−259.0	−0.2
−308.5	−308.4	−0.1	−278.9	−278.9	0	−255.5	−255.7	0.2
−307.6	−307.8	0.2	−278.2	−278.2	0	−254.4	−254.6	0.2
−305.4	−305.5	0.1	−277.4	−277.5	0.1	−253.6	−253.6	0
−292.5	−292.4	−0.1	−276.1	−276.1	0	−252.8	−252.9	0.1
			−274.2	−274.4	0.2	−251.8	−251.7	−0.1
			−272.8	−272.9	0.1	−250.8	−250.9	0.1
			−272.1	−272.1	0	−249.3	−249.3	0
			−271.3	−271.4	0.1	−248.5	−248.5	0
			−270.5	−270.6	0.1	−247.5	−247.7	0.2
			−269.8	−269.8	0	−246.8	−246.8	0
						−246.1	−246.1	0
						−245.3	−245.3	0
						−244.5	−244.5	0

In Fig.4 the eight experimental spectra and the calculated energy of Stark manifolds are combined together. More systematic accordance with each other is demonstrated. The field interaction increases as the applied field varies from 0 to 600 V/cm, the initial degenerated energy levels separate from each other by free-degeneracy induced by external fields. Also Stark expansion and redistribution of atoms cause the intensity of spectra to weaken..

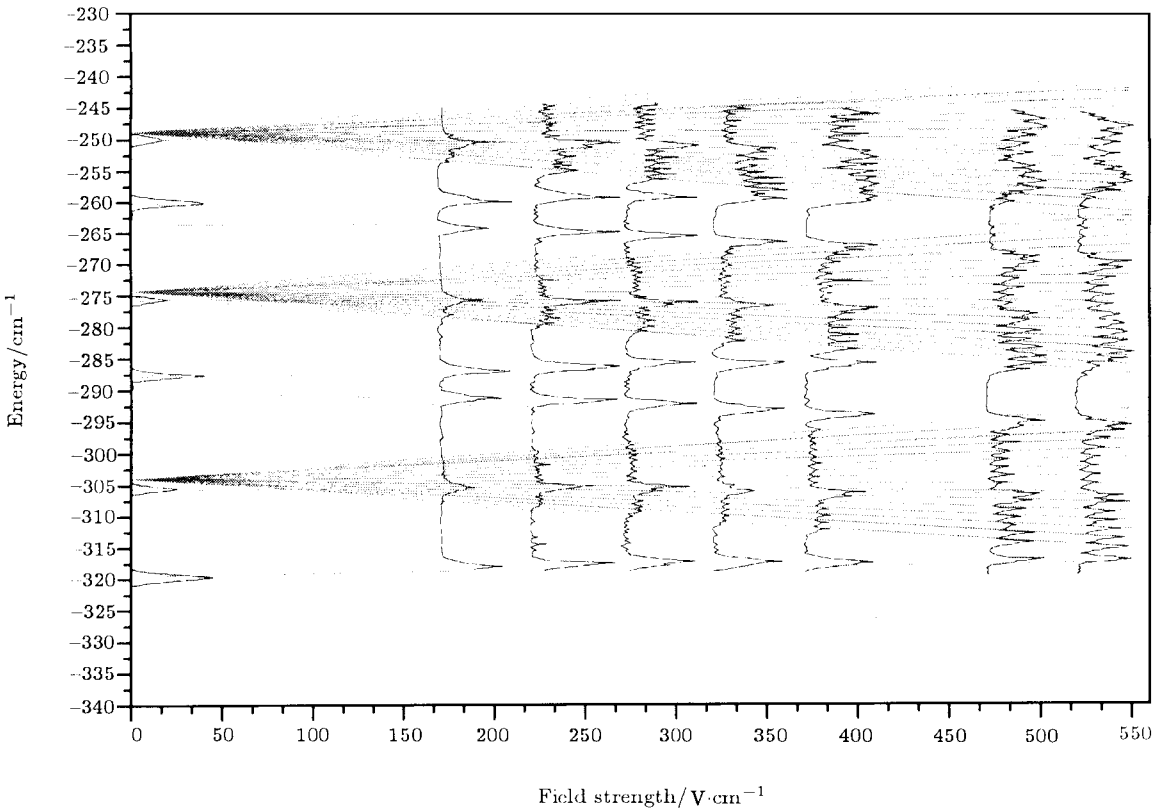


Fig.4. Cs Stark structure

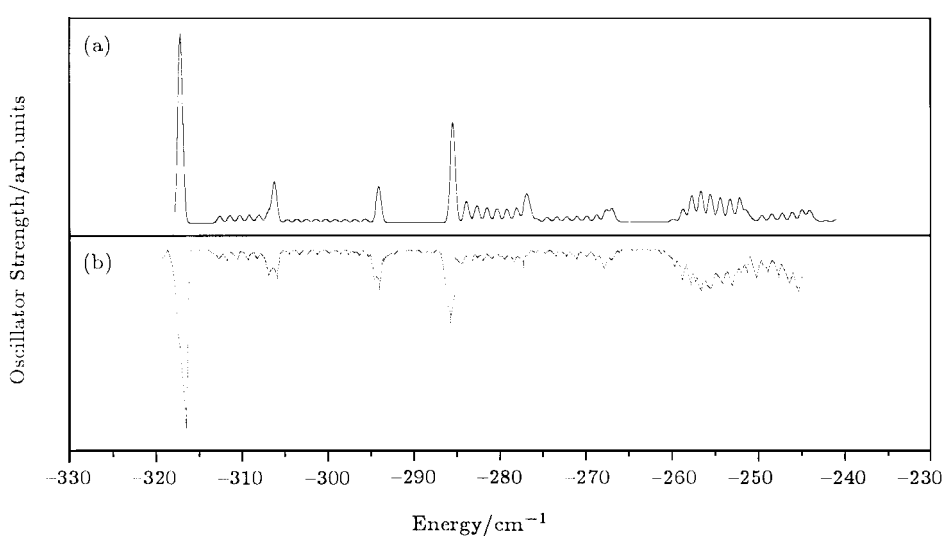


Fig.5. Oscillator Strength: Curve (a) is the calculated distribution; Curve (b) is the fitted from experimental spectrum in the same field of 420V/cm

Unlike hydrogen atoms in external electric fields, the Stark separation of Cs Rydberg atoms first occurs from S-portion of the manifold of n^2S because of the large quantum defect and strong coupling between Stark sublevels in low fields, when in high fields the edge angular momentum states of Stark fan structure begin to couple with the states n^2D and $(n+1)^2$D and make transition spectra tend to be homogeneous, so n-mixing starts. The oscillator strength is also computed in our case to confirm the agreement, and we find the same results as obtained from the above (see Fig.5). Curve (a) in Fig.5 was obtained from fitting of the experimental spectrum in a field of 420V/cm by peaks divided by normalized area, and curve (b) obtained from the PM theoretical calculation. But it is noticeable that the discrepancy exists between them in larger n or high energy level and we can attribute it to the saturation effect in experiment and errors.

We can conclude that for the Stark structure of Cs Rydberg atoms in the vicinity $n^* = 21 - 23$ the result calculated theoretically by PM method is in excellent agreement with that of experimental measurement within permissible experimental errors, which may be attributed mainly to three factors: 1) the 2nd pulse laser linewidth about 12GHz, 2) nonlinear scanning of the second laser, and 3) transitions between $m_l \neq 0$ states caused by incomplete line polarization of lasers. More subtle and precise measurement is required for further research.

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