Relativistic J-Dependence of the Isotope Shift in the 6s-6p Doublet of Ba II

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Abstract: The collinear laser-ion beam technique has been used to measure the isotope shift and hyperfine structure in the 6s-6p doublet (4934 Å, 4554 Å) of Ba II for all seven stable isotopes. The influence of the excited $^2P_{1/2}$ and $^2P_{3/2}$ states on the field shift leads to a difference of 2.5(3) % in the electronic F factors. The specific mass shifts differ by $\{A-A'\}$ 2.3(3) MHz which corresponds to about 12 % of the normal mass shift.

1. Introduction

Barium has been among the favoured candidates for testing new methods of laser spectroscopy. Thereby, most of the low-lying atomic (Ba I) and ionic (Ba II) energy levels have been studied with high resolution. In the alkali-like spectrum of singly-ionized barium the hyperfine splitting of the 6s $^2S_{1/2}$ ground state is very precisely known from optical pumping microwave resonance experiments [1,2]. Fast ion beam-laser techniques using quantum beats [3] or hole burning by velocity-selective optical pumping [4] have yielded the hyperfine structure of the excited 6p $^2P_{1/2}$ and 6p $^2P_{3/2}$ states. These levels were also involved in the collinear-beam and crossed-beam studies of the isotope shift and hyperfine structure in the 5d-6p transitions from the metastable 5d doublet ([5] and references therein). The improved accuracy of the latest experiment has revealed a J-dependence of the isotope shift of the 5d $^2D_{3/2}$ and 5d $^2D_{5/2}$ levels.

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Here, we report the high-resolution measurement of the isotope shift in the 6s-6p resonance doublet (4934 Å, 4554 Å) for all seven stable isotopes of barium. Earlier results of Doppler-limited optical spectroscopy [6,7] are improved, particularly in the transition $6s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$, and complemented by measurements on the least abundant isotopes.

The scope of this work is twofold:

(i) A comparison of the isotope shift in both components should demonstrate the different influence of the $6p\ ^2P_{1/2}$ and $6p\ ^2P_{3/2}$ wave functions on the field and mass shift.

(ii) The semi-empirical evaluation of the nuclear mean square charge radii $\delta\langle r^2 \rangle$ is preferably based on the resonance transition in the alkali-like Ba II system, for which the electronic F factor is calculated from either the hyperfine splitting of the 6s electron or the Goudsmit-Fermi-Segrè-formula with identical results [8,9]. This conventional procedure has been adopted in the recent work on stable, neutron-deficient and neutron-rich barium isotopes ($122 \leq A \leq 146$) [10,11] for which the isotope and isomer shifts were measured in the Ba I resonance line $6s^2\ ^1S_0 - 6s6p\ ^1P_1$ (5536 Å) and related to the Ba II line $6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$ (4934 Å) via a King-plot. The present measurement should yield more accurate Ba II data and show to what extent the non-vanishing electron density at the nucleus from the $p_{1/2}$ electron has influenced the results.

2. Experiment

A schematic view of the experimental set-up is given in Fig. 1. The barium ions are provided by a surface-ionization source and accelerated to an energy of about 50 to 60 keV. After mass separation in a magnetic field the ion beam is deflected to overlap coaxially with the beam of a single-frequency cw dye laser. The laser is operated using the dyes Stilben 3 (4554 Å) or a mixture of Coumarin 2 and Coumarin 102 (4934 Å), pumped by the UV lines of an argon ion laser. For Doppler-tuning of the effective laser frequency via the kinetic energy of the ions, the interaction region is at a variable potential of $\pm 10\%$ of the main acceleration voltage. The post-acceleration/deceleration system in front of this part of the apparatus is designed as to avoid an influence on the ion-beam divergence. The change of the kinetic energy also prevents optical pumping of the atoms before they enter into the observation region. Resonance excitation within the observation length of 20 cm is detected by counting the

fluorescence photons with an efficiency of 0.5 %, including the 20 % quantum efficiency of the photomultiplier. For background-free observation the 35 % decay branch to the metastable 5d levels (6496 Å, 6141 Å and 5853 Å) can be selected by optical filters. However this does not improve the signal to noise ratio, because the stray-light background is low compared to the total fluorescence yield for the typical beam intensities between 10 pA and 10 nA. The count rates from the fluorescence photons are stored in a mini-computer as a function of the post-acceleration voltage. For an accurate determination of isotope shifts and hyperfine splittings this voltage as well as the main acceleration voltage is measured to an accuracy of 10^{-4} by precision voltage dividers and digital voltmeters. Drifts of the main acceleration voltage are eliminated by an active stabilization of the 60 kV power supply [11,12].

Typical resonance curves are shown in Fig. 2. One measuring cycle includes alternating scans on three isotopes to minimize effects of long term voltage or laser drifts. The experimental line widths is about 30 MHz. It is thus somewhat broader than the natural line widths of 21 MHz (4934 Å) and 25 MHz (4554 Å), respectively, due to the contribution of the residual Doppler width and some power-broadening. The isotope shifts and hyperfine splittings are calculated from voltage differences via the relativistic Doppler equation:

$$\Delta v^{1,2} = v_0 \left\{ \left(\frac{1 - \beta_2}{1 + \beta_2} \right)^{1/2} - \left(\frac{1 - \beta_1}{1 + \beta_1} \right)^{1/2} \right\} \quad (1)$$

with $\beta_i = \sqrt{2eU_i/m_i}$. The relative line positions are determined by application of a fitting routine. Due to the different line width contributions, the line profile is a complex function which can be well described by the convolution of a Lorentzian and a Gaussian shape.

3. Results

The results for the isotope shifts of all stable barium isotopes, relative to ^{138}Ba , are given in Table 1. The errors quoted include the experimental systematic uncertainties, mainly due to the high-voltage calibration, and the statistical standard deviation from the fitting procedure. These data for the first time include the distinct observation of the difference in isotope shift between both fine structure components of the resonance doublet in an alkali-like system.

The hyperfine structure parameters A and B of the two odd isotopes, quoted in Table 2, are in good agreement with the earlier and partly more accurate data obtained by various techniques [2,3,4,13]. Becker et al [13] discussed these parameters, in particular with the aim of extracting nuclear quadrupole moments from the B-factors of the $6p\ ^2P_{3/2}$ state.

4. Discussion of the Isotope Shifts

For a given transition i the isotope shift between the isotopes A and A' is the sum of the nuclear-shape dependent field shift and the mass shift [8] :

$$\delta\nu_i^{A,A'} = F_i \delta\langle r^{-2} \rangle^{A,A'} + M_i \{ (A'-A)/(A'A) \} \quad (2)$$

By convention, the mass shift constant $M_i = N_i + S_i$ is separated into the trivial normal mass shift constant $N_i = \nu_i/1836.1$ and the specific mass shift constant S_i , which accounts for electron-correlation effects. The J-dependence of both electronic factors F_i and S_i , can be separated via the procedure of King. In Fig. 3 we have plotted $\delta\nu_i \{ (A'A)/(A'-A) \} - N_i$ for the transition $6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$ (4934 Å) against the same quantity for the second transition $6s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$ (4554 Å). The straight line through the data point has the slope

$$\rho = F(4934\text{ Å}) / F(4554\text{ Å}) = 0.975 \quad (3)$$

and the intercept

$$S(4934\text{ Å}) - \rho S(4554\text{ Å}) = 40\text{ (5) GHz.}$$

Thus the field shift of the transition $6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$ is $\sigma = (1 - p) = 2.5\ (3)\ \%$ smaller than that of the transition $6s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$. This small difference in the field shift of the doublet components can be ascribed to the relativistically non-vanishing charge density of the $p_{1/2}$ electron at the origin. This charge density is produced by the admixture of the small Dirac component, which has the radial dependence of a s-wave function. Fradkin [14] and Ionescu-Pallas [15] used hydrogenic wave functions to estimate this effect and obtained:

$$\sigma_H = \alpha^2 Z^2 / \{ 1 + \sqrt{1 - \alpha^2 Z^2} \}^2 \quad (3)$$

For the present case of $Z = 56$, this simplified approach gives $\sigma = 4.6\ \%$ and thus overestimates the effect by almost a factor of 2. The discrepancy is obviously due to the omission of the effects of screening of the core electrons on the $^2S_{1/2}$ and $^2P_{1/2}$ wave functions. In a semi-empirical description of the alkali level structure, this effect is accounted for by the introduction of effective charges Z_i and Z_a and effective quantum number n^* into the hydrogenic formulae. For the valence-electron density at the nucleus, the hydrogenic expression Z^3/n^3 has to be replaced by $Z_a^2 Z_i / n^{*3}$ (Goudsmit-Fermi-Segrè-formula) [9]. This approach has been successful in reproducing the interaction constants of fine structure, hyperfine structure and isotope shift. All these are due to the central part of the electron wave function close to or within the nucleus, which is also true for σ . We therefore can correct formula (3) to

$$\sigma = \sigma_H \{ Z_i(p_{1/2}) / n^{*3}(p_{1/2}) \} / \{ Z_i(s_{1/2}) / n^{*3}(s_{1/2}) \} \quad (4)$$

With the conventional assumption $Z_i(p_{1/2}) = Z - 4$, $Z_i(s_{1/2}) = Z$, and taking the n^* values from the experimental term energies T via the Rydberg-Ritz formula $n^* = Z \sqrt{R_\infty/T}$, one obtains $\sigma = 2.7\ \%$ in good agreement with the experiment. Core polarization and the admixture of higher p states should have only little influence on the ratio σ . The configuration mixing can be estimated by the method of Fermi [16]. It yields about 5 % admixture of higher np $^2P_{1/2}$ states to the 6p $^2P_{1/2}$ wave function. The corresponding reduction of the 6p $^2P_{1/2}$ charge density and thus the relative influence to the ratio σ will therefore not exceed 5 %. As Fischer [17] points out, the core polarization is of the order of 8 % for the barium 6p states and influences both the $^2P_{1/2}$ and $^2P_{3/2}$ levels in the same manner without contributing to a difference in the isotope shift.

Ab initio calculations with multi-configuration Dirac-Fock wave functions have recently been carried out by Fricke and Rosén [18] for the electron-charge densities and the electronic factors F_i in a number of Ba I and Ba II transitions. The F_i ratios can be directly compared with the experimental data. For the present case one obtains from $F(4934 \text{ Å}) = 3.80 \text{ GHz/fm}^2$ and $F(4554 \text{ Å}) = 3.88 \text{ GHz/fm}^2$ the ratio $\rho = 0.979$ which is very close to the experimental value $\rho = 0.975(3)$. Although this confirms that the F_i ratios are rather well reproduced by the new calculations, it is difficult to draw conclusions about the accuracy of their absolute values and their reliability for the evaluation of $\delta\langle r^2 \rangle$ in a sequence of isotopes (cf. discussion in Ref. [11]).

The specific mass shift constants S_i of the two transitions differ by 40(5) GHz (because $\rho \approx 1$), i.e. the specific mass shifts differs by $\{A'-A\} 2.3(3) \text{ MHz}$. This value corresponds to about 12 % of the normal mass shift and is therefore in accordance with the qualitative assumption that $|S_i| \leq N_i$ for all alkali-like s-p transitions [8]. To date no quantitative calculations have gone beyond this accuracy due to the problem of calculating the small specific mass shifts as differences of the much larger individual-level shifts. The non-relativistic specific mass shift operator depends only on the orbital angular momentum L [19] whereas differences within a fine-structure multiplet are introduced by relativistic corrections. A theoretical formalism of these corrections has been developed by Stone [20] but no quantitative results are available for the levels considered here.

5. Conclusion

Although the small difference in the isotope shifts due to the np $^2P_{1/2}$ and np $^2P_{3/2}$ wave functions is of minor importance in the nuclear physics interpretation of isotope shift data from s-p transitions, it is a clean example for quantitative comparison of experimental results with semi-empirical as well as ab initio calculations. Earlier data are available only for the $6s^26p - 6s^27s$ doublet in Tl I, where the difference in the total isotope shift is 10(5) %, corresponding to 24 % of the contribution of the 7s electron ([17] and references therein). Assuming $|\psi_0|^2_{6s} / |\psi_0|^2_{7s} = 8$ [17] one derives from this measurement $\sigma = 3(1) \%$ in agreement with our semi-empirical estimation, which yields $\sigma = 4 \%$. Transitions analog to

the ones studied here, such as the 6s-6p doublet in Cs I and the 7s-7p doublet in Fr I and Ra II should be accessible for further experiments. In all these cases, only one component of the resonance doublet has so far been studied by high-resolution spectroscopy [21,22,23].

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Table 1: Isotope shifts in the 4554 Å and 4934 Å line of Ba II, relative to the isotope ^{138}Ba .

$\delta_{\nu}^{138,A}$ (MHz)				
4934 Å			4554 Å	
A	This work	Ref. #[6], †[7]	This work	Ref. [6]
138	0	0	0	0
137	271.1(1.7)	283.0(6.9)# 264(30)†	279.0(2.6)	285(30)
136	179.4(1.8)	186.5(7.3)# 168(12)†	186.9(2.1)	177(15)
135	348.6(2.1)	357.4(7.5)# 378(30)†	360.7(2.2)	381(30)
134	222.6(3.0)	229.0(7.0)# 243(12)†	233.9(3.7)	249(12)
132	278.9(4.0)		294.9(4.2)	
130	355.3(4.4)		372.3(4.9)	

Table 2: A and B factors of the levels under study.
All values in MHZ.

	^{135}Ba		^{137}Ba	
	This work	Ref. †[2], †[13], #[3], ϕ[4]	This work	Ref. †[2], †[13] #[3], ϕ[4]
$A^2S_{1/2}$	3593.3(2.2)	3591.67011718(24)†	4020.3(2.3)	4018.87083385(19)†
$A^2P_{1/2}$	665.0(1.6)	663.8(1.7)†	744.1(1.6)	742.3(1.3)†
$A^2P_{3/2}$	113.2(0.8)	114.6(1.0)† 112.72(32)# 113.1(0.5)ϕ	126.7(1.1)	125.5(0.6)† 125.95(35)# 125.8(0.5)ϕ
$B^2P_{3/2}$	59.4(2.3)	67.5(7.8)† 60.45(35)# 60.7(1.2)ϕ	95.0(3.7)	97.1(6.6)† 92.53(40)# 91.6(1.2)ϕ

6. Figure captions:

Figure 1: Schematic view of the experimental set-up for collinear fast beam-laser spectroscopy on ions.

Figure 2: Resonance curves for the isotopes ^{130}Ba , ^{135}Ba and ^{138}Ba . in the transition $6s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$ (4554 Å). The F values of the excited and ground state sublevels are given at the corresponding hyperfine components of ^{135}Ba .

Figure 3: King-plot between the 4554 Å and the 4934 Å line. The inserts show the data points and the origin enlarged by a factor of 10.

Fig. 1

Fig. 2

Fig. 3