Fine structures of the $nd\ ^2D$ and $nf\ ^2F$ sequences in singly ionized beryllium

B. Denne,* H. Dickow, and O. Poulsen
Institute of Physics, University of Aarhus, DK-8000 Aarhus C, Denmark
(Received 2 June 1980)

Fine structures in the $nd\ ^2D$, $n = 4,5$ and $nf\ ^2F$, $n = 4,5,6$ levels in Be II have been measured, using fast-beam, level-crossing spectroscopy. The results, accurate to within 10^{-3}, point to small deviations from a hydrogenic fine-structure splitting due to correlation effects involving the $1s^2$ core for the $nf\ ^2F$ levels.

INTRODUCTION

The study of fine and hyperfine structures in Rydberg states, in particular, in quasihydrogenic atoms, has revealed many properties, among others the inverted $nd\ ^2D$ fine structure in the alkalilike atoms. This feature has been explained theoretically[1,2] as stemming from a strong exchange-core polarization of the p core induced by the Rydberg electron. Also, the $nf\ ^2F$ excited states show deviations from a pure hydrogenic fine-structure splitting.[2] This deviation is, in turn, explained as an exchange core-polarization effect, the deviation becoming larger for large n and even yielding inverted fine-structure intervals for the heavy alkalis. As pointed out by Cooke et al.,[3] the Li I isoelectronic sequence offers good possiblilites for testing the validity of the core-polarization contributions. Thus the deviations from hydrogenlike fine-structure (fs) splittings in the Li I isoelectronic sequence must be attributed to correlation effects involving s-type excitations in the core.

Such effects give very strong contributions to the hyperfine structure hfs of the $2p\ ^2P$ level in the Li I isoelectronic sequence.[4,5] In Li I, the hfs of the $2p\ ^2P_{3/2}$ level is inverted, whereas in Be II, the hfs is close to zero with the polarization and correlation effects giving contributions which nearly cancel the one-electron direct contribution. In Li I, the situation looks more normal concerning the fs of the $(1s)^2nl$ configurations. The $n = 3, 4\ ^2D$ levels have been studied, using the level-crossing (LC) technique,[6,7] whereas the $n = 7 - 10\ ^2D$, ^2F, and ^2G fs intervals were measured,[3] using a microwave resonance technique. Within the quoted uncertainties, these authors do not find any deviations from a hydrogenic fs splitting, although the experimental results are all slightly lower than the hydrogenic splittings.

In order to further test the possibility of deviations from hydrogenlike behavior in such a simple alkalilike system, we have performed LC measurements in the beryllium ion (Be II). The fast-beam level-crossing technique, which provides a convenient tool for studying ionic systems, was applied. The LC technique is Doppler-free, with a resolution only limited by the natural width of the level studied. In the present work, the fs splittings are determined to within 10^{-3}. By comparing the experimental splittings with the calculated hydrogenic intervals, this precision allows the establishment of a nonhydrogenic behavior for the $4f\ ^2F$ level, whereas the $nd\ ^2D$ levels are found to be almost purely hydrogenic.

These fine structures have previously been studied by conventional spectroscopy[8] and by zero-field quantum beats,[9] the precision being 3×10^{-2}. The $5f\ ^2F$ level has previously been studied with a technique similar to the one used in this work.[10]

EXPERIMENTAL

The experimental apparatus has been described previously[11] and only the small changes will be discussed. The 80-kV isotope separator at the University of Aarhus was used to produce a beam of Be^+ ions, which were excited in a helium-filled gas cell, placed inside the pole gap of an electromagnet. The ion beam, the magnetic field, and the observation direction were at right angles to one another.

The magnetic field was swept by a current supply which was stabilized on a linearized Hall probe. The magnetic field was calibrated using a rotating-coil gaussmeter, which, in turn, was calibrated against NMR. Taking into account the inhomogeneities of the field in the pole gap, the magnetic field is determined to within 0.5 gauss in the interaction region. The motional Lorentz force, perpendicular to the fast beam as well as to the magnetic field, was eliminated by applying a high voltage to a set of deflection plates. Both the high-voltage amplifier and the Hall-stabilized current supply were computer interfaced.

The scattered light was polarization-analyzed and dispersed in a fast (f 3.5) spectrometer (slits parallel to the ion beam) equipped with a

Peltier-cooled EMI 6256S photomultiplier. The data were then stored for subsequent analysis.

RESULTS

The main limitations of the fast-beam LC method are the requirements of sufficient population as well as of anisotropic-excitation cross sections. The $nd\,^2D$, $n=4,5$, and $nf\,^2F$, $n=4,5,6$, have previously been studied,[10] and the above two criteria were known to be fulfilled. At 50 keV of incident beam energy, anisotropies of the order of 15% are found for all of the levels studied. To ensure internal consistency, at least two LC's were recorded for each level. In Fig. 1 is shown a typical result. The resonance positions were found by a least-squares adjustment of a single Lorentzian. Before we evaluate the fs splittings by means of the Breit-Rabi formula, we shall consider the systematic errors. Among the most serious systematic-error sources are the Stark shifts in the residual motional field. Also, the nonzero hyperfine interaction as well as the quadratic Zeeman effect will be considered.

A. Stark shift

The effect of this motional electric field, if not properly compensated for, is not only a shift of the energy levels, and thus of the level crossings, but also a coupling of the two crossing levels, converting a level crossing into an anticrossing. Calculating the shift within a single LS multiplet, we get

$$\langle \alpha SLJM \,|\, \mathcal{H}_{st} \,|\, \alpha SLJ'M' \rangle = \sum_{k=0,2;q} (H_s)^k_q (-1)^{J-M+L+S+J'} [(2k+1)(2J+1)(2J'+1)]^{1/2} \begin{Bmatrix} L & J & S \\ J' & L & 2 \end{Bmatrix} \begin{pmatrix} J & k & J' \\ -M & q & M \end{pmatrix}, \tag{1}$$

with $\mathcal{H}_{st} = -e\vec{r}\cdot\vec{\mathcal{E}}$ being the perturbation in the electric field $\vec{\mathcal{E}}$. As only the quadrupolar part gives rise to a relative shift of the various levels, only $(H_s)^2_0$ need be evaluated if we choose $\vec{\mathcal{E}} = (0,0,\mathcal{E})$,

$$(H_s)^2_0 = -e^2\mathcal{E}^2\sqrt{\tfrac{2}{3}} \sum_{\beta L'} \frac{l_{\max}\mathfrak{R}^2}{E(\alpha LS) - E(\beta' L'S)} \begin{Bmatrix} 1 & 1 & 2 \\ L & L & L' \end{Bmatrix}. \tag{2}$$

Here $\mathfrak{R}$ is the one-electron integral

$$\mathfrak{R} = \int_0^\infty rR(nl)R(n'l-1)dr, \tag{3}$$

which can be computed using hydrogenlike wave functions. This procedure is justified based on the regularity of the energy levels and quantum defects in the nd and nf series in Be II.[8]

In Eq. (1), the summation, which extends over all opposite parity levels $\beta L'$, is limited to one n manifold because of the quasihydrogenic nature of the atom.

However, the experimental conditions correspond to the electric field $\vec{\mathcal{E}}$ being perpendicular to both the ion beam $\vec{v} = (0, v, 0)$ and the magnetic field $\vec{H} = (0, 0, H)$. Thus a rotation of $\pi/2$ around the y axis will bring the electric field down onto the x axis, thus representing the experimental reality. $(H_s)^2_0$ will then be transformed into an $(H'_s)^2_0$ and an $(H'_s)^2_{\pm 2}$ part, the former giving rise to a shift of the crossing energy levels, whereas the $(H'_s)^2_{\pm 2}$ part converts the LC between a pair of magnetic sublevels, with $\Delta M = \pm 2$, into an anticrossing. In this work we have eliminated this motional electric field to be less than 50 V/cm in order to reduce the uncertainty related to the calculation of the radial integrals $\mathfrak{R}$. The crossing positions are thus changed less than 0.01 gauss for the first-level crossing $(7/2, -7/2) - (5/2, -3/2)$ in the $4f\,^2F$ level, in a field of 1 kV/cm. For all levels studied, the resonances were found to be pure Lorentzians, which is clear evidence of the effective elimination of the Stark shifts.

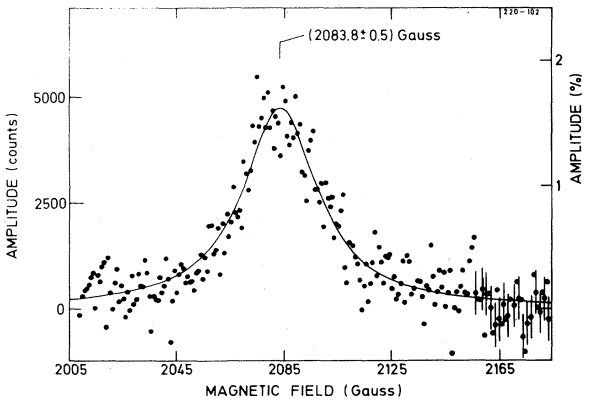

FIG. 1. The first-level crossing $(\frac{5}{2}, -\frac{5}{2}) - (\frac{3}{2}, -\frac{1}{2})$ in $4d\,^2D$ in ^{9}Be II. The FWHM of (35 ± 3) gauss yields a $4d$ lifetime of (2.8 ± 0.3) ns, in agreement with previous work (Ref. 10).

B. Hyperfine structure

Beryllium has a nuclear spin of $\frac{3}{2}$. The hyperfine structure can be calculated using hydrogenic wave functions. For all levels studied, the result is[9] a hyperfine splitting of less than 5% of the nat-

ural linewidth. Thus the nuclear and electronic spins are effectively uncoupled. Hence, each crossing consists of four unresolved Lorentzians with a center of gravity close to the single-crossing position for the case of a vanishing hyperfine structure. The FWHM of these resonances is equal, thus justifying our analysis using only one Lorentzian.

C. Quadratic Zeeman effect

The quadratic Zeeman effect, proportional to the square of the vector potential,[12] introduces a shift of the crossing energy levels. Using hydrogenlike wave functions to calculate the average $\langle r^2 \rangle$, these shifts are, in a magnetic field H, given by

$$\Delta E = \frac{e^2H^2}{8mc^2} \frac{n^2[5n^2+1-3l(l+1)]}{2Z^2} \frac{m_J^2+J^2+J}{2J(J+1)} \qquad (4)$$

for a level $|nslJm_J\rangle$ with an effective nuclear charge Z_{eff}. These energy shifts result in shifts of the crossing positions smaller than 0.02 gauss for all the levels studied.

The fs intervals are now evaluated by diagonalizing the Hamiltonian

$$\mathcal{H} = \mu_B H(g_l L_Z + g_s S_Z) + \delta W, \qquad (5)$$

where μ_B is the Bohr magneton and g_l and g_s are the g factors for the orbital and the spin motions. We have used $g_l = 1$ and $g_s = 2.0023$. δW is the fine-structure splitting. Prior to diagonalizing $\mathcal{H}$, the experimentally found level crossings are corrected for the systematic errors discussed. The final results are shown in Table I. The uncertainties include systematic as well as statistical errors. Also shown in Table I are the hydrogenic fine-structure splittings calculated by means of

$$\delta W(\text{hy}) = \frac{R\alpha^2 Z_{\text{eff}}^4}{n^3 l(l+1)}, \qquad (6)$$

where n is the main quantum number and $Z_{\text{eff}} = 2$ is the effective nuclear charge for Be^+. Using the effective quantum numbers n^* instead of the integer main quantum numbers[8] does not change the calculated fine structures for the nf levels as they show quantum defects less than 0.0003, whereas for the $4, 5d\,^2D$ levels, quantum defects of 0.0021 and 0.0022 change the calculated fine structures 11 and 5 MHz, respectively, toward larger splittings.

The $3d\,^2D$ level, which decays at 1512 Å, was not measured because an electric field of 6 kV/cm was needed to compensate for the Lorentz field.

TABLE I. Measured and calculated hydrogenic δW(hy) fine-structure intervals in Be II (MHz).

Level	δW	δW(hy)
$4d\,^2D$	7293 ± 4	7299
$5d\,^2D$	3730 ± 10	3737
$4f\,^2F$	3668 ± 5	3649
$5f\,^2F$	1870 ± 5	1868
$6f\,^2F$	1077 ± 5	1081

This high electric field made it difficult to transport the beryllium-ion beam into the interaction volume due to strong fringing fields at the entrance to the pole gap.

The previous LC measurements[10] of the $5f\,^2F$ level deviate 3% from this more precise measurement, the reason being the much improved magnetic-field monitoring equipment as well as compensation of the motional electric field.

DISCUSSION

The gross features of the spectra in the lithium-isoelectronic sequence are hydrogenlike with small quantum defects, in particular, for the high angular momentum states. The fine structures of the low-lying levels were known from classical spectroscopy[8] to be normal and close to hydrogenic fine structures. Previous works[3,6,7] found the fine structures in the d, f, and g levels in neutral lithium to be hydrogenic to within 0.5%.

From Table I it is seen that the deviations found in this work for the beryllium ion are hydrogenic to within 10^{-3} for the $nd\,^2D$, $n = 4, 5$ levels. Using the effective quantum numbers, this deviation is increased to 2×10^{-3}. Also for the $nf\,^2F$, $n = 4, 5, 6$ levels, only small deviations are found. However, with the precision obtained in the present study, deviations occur. For the $4f\,^2F$ level, we find an fs splitting larger than the hydrogenic splitting, whereas slightly smaller values are found for the $5, 6f\,^2F$ intervals. With the p electrons in the core eliminated, this deviation must be due to correlation effects. Such effects are expected to give only small contributions, and the deviations found in this work amount to only 0.4% at most. Thus the present study confirms the explanation that the nature of the inversion of the fine structure in the alkali atoms with p cores is due to exchange-core polarization, at the same time pointing out that small deviations from a hydrogenic fs are due to correlation effects involving the $(1s^2)$ core.

*Permanent address: Department of Physics, University of Lund, S-22362 Lund, Sweden.

[1]L. Holmgren, I. Lindgren, J. Morrison, and A.-M. Mårtensson, Z. Phys. A 276, 179 (1976).
[2]R. M. Sternheimer, J. E. Rogers, and T. P. Das, Phys. Rev. A 17, 505 (1978).
[3]W. E. Cooke, T. F. Gallagher, R. M. Hill, and S. A. Edelstein, Phys. Rev. A 16, 1141 (1977).
[4]S. Garpman, I. Lindgren, J. Lindgren, and J. Morrison, Z. Phys. A 276, 167 (1976).
[5]O. Poulsen, P. S. Ramanujam, and D. B. Iversen, J. Phys. B 8, L450 (1975).
[6]K. Fredriksson, H. Lundberg, and S. Svanberg, Z. Phys. A 283, 227 (1977).
[7]R. L. Smith and T. G. Eck, Phys. Rev. A 2, 2179 (1970).
[8]L. Johansson, Ark. Fys. 20, 489 (1961).
[9]O. Poulsen and J. L. Subtil, Phys. Rev. A 8, 1181 (1973).
[10]O. Poulsen, T. Andersen, and N. Skouboe, J. Phys. B 8, 1393 (1975).
[11]T. Andersen, S. Isaksen, D. B. Iversen, and P. S. Ramanujam, Phys. Rev. A 18, 1079 (1978).
[12]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Springer, Berlin, 1957), p. 227.