Time-differential level-crossing g-value measurements of the $6p\,^2P$ fine-structure levels in ^{138}Ba II using an optical-induced orientation or alignment of a fast ionic beam

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A time-differential level-crossing technique using laser excitation of a fast ion beam is presented. The fast beam thus oriented or aligned makes possible measurements of the Landé g factors. They have been measured to be $g_{1/2} = 0.672 \pm 0.006$ and $g_{3/2} = 1.328 \pm 0.008$ for the $6p\,^2P_J$ levels in ^{138}Ba II.

INTRODUCTION

The classical level-crossing technique[1] has been used to obtain a variety of information such as the Landé g factors and fine and hyperfine structures of excited states. Pulsed excitation, where electrons [2] have been used, carbon-foil excitation of a fast ion beam,[3] or fast beam-gas excitation,[4] yield the same type of information as does modulated excitation with electrons[5] or light.[6] The extension of these techniques to fast ion beams,[3,4,7] where collisional excitation was applied, made these classical level-crossing-type experiments applicable to the study of highly excited singly and doubly ionized atoms. Laser excitation of fast ion beams has made possible cascade-free lifetime[8-11] and hyperfine-structure[12] determinations, as has also pulsed-laser excitation of atoms in a vapor.[13]

In this work, a time-differential level-crossing technique by means of laser excitation of a fast ion beam is presented. It is shown that a high degree of orientation and alignment can be induced into the excited beam ions, thus making possible measurements of fine and hyperfine structures and Landé g factors. The g values of the $6p\,^2P_J$ levels in ^{138}Ba II have been determined in the present experiment and are found to be in good agreement with the classical Zeeman spectroscopic values.[14]

THEORY

In the present work, a cw dye laser with a spectral half-width of 0.3 Å has been used to excite a 50-keV ion beam of $^{138}Ba^+$ with a beam divergence corresponding to 0.2 Å. Figure 1 shows a part of the Ba II level relevant to this study. The metastable $5d\,^2D_J$ levels are created in the ion source of the ion accelerator used. The intensity expressions in the following will assume the laser beam to be parallel, an assumption which is justified because the depth of field is calculated to be several centimeters. The exciting laser profile $L(x)$ seen by the ion traversing it is established experimentally. At the laser powers used (typically 20–100 mW) no saturation or pumping effects have been taken into account. Based on the interaction time t of the laser and the ions, the spectral half-width of the laser, and the lifetime of the excited state and its branching ratios to the ground and metastable states, it has been estimated that about 10–20% of all metastable ions are excited once but less than 1% of them twice.

The fluorescent light emitted from the $6p\,^2P_J$ levels are measured as a function of an external magnetic field at fixed time t_0 after excitation. With a beam velocity of v (mm/nsec), the time t_0 can be converted directly into distance x_0 along the ion beam as $x = vt$.

Now let $i(x_0, H)$ describe the fluorescent light emitted at time $t_0 = x_0/v$ after excitation. The total intensity detected will then be given by

$$I(x_0, H) = \int_{-\infty}^{\infty} \int L(x) N(x) i(x - x', H) D(x') dx\, dx'$$

$$= \int i(x, H) P(x)\, dx, \qquad (1)$$

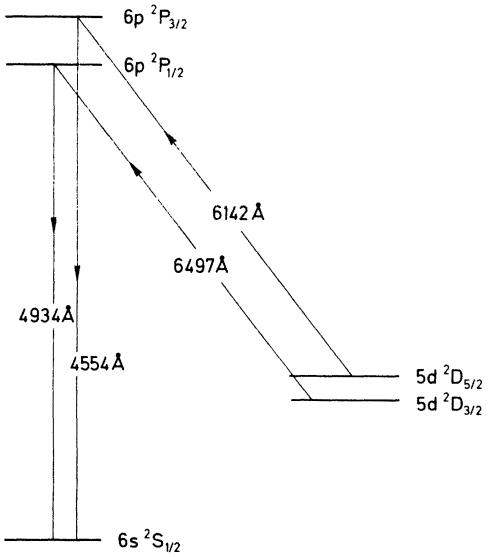

FIG. 1. Partial Ba II level scheme.

where

$$P(x) = \int D(x')N(x+x')L(x+x')\,dx' \tag{2}$$

is an effective experimental profile consisting of the exciting laser profile $L(x)$, detection profile $D(x)$, and a function $N(x)$, taking into account the ion-beam divergence. $N(x)$ is given by

$$N(x) = (a - x\sin\varphi)^2 , \tag{3}$$

where a^2 is the area of the two square apertures placed symmetrically around the ion-beam focus at distance c, thus defining the ion beam; φ is the divergence angle of the ion beam, defined as $\tan\varphi = a/c$.

The effect of $P(x)$ on an exponential decay is only that of a multiplicative constant when detection is performed outside the exciting profile. When dealing with a cosine describing the time-resolved precession of the excited state in the magnetic field, a damping of the oscillations is introduced. Except for the influence from $N(x)$, which changes the average time t_0 spent by the excited ions in the magnetic field (typically by 0.1–0.3%), $P(x)$ does not change the frequency of the cosine.

The fluorescent intensity $i(x,H)$ is given by the time-differential version of the Breit equation,

$$\begin{aligned} i(x,H,\hat{e}_1,\hat{e}_2) = \sum_{JJ'FF'MM'} \exp\left[\left(\frac{-i[E(JFM)-E(J'F'M')]}{\hbar} - \Gamma\right)\frac{x}{v}\right] \\ \times \sum_{J_0F_0M_0} \langle JFM|\hat{e}_1\hat{d}|J_0F_0M_0\rangle \rho(J_0F_0M_0)\langle J_0F_0M_0|\hat{e}_1^*\hat{d}^\dagger|J'F'M'\rangle \\ \times \sum_{J,F,M} \langle J'F'M'|\hat{e}_2\hat{d}|J_1F_1M_1\rangle\langle J_1F_1M_1|\hat{e}_2^*\hat{d}^\dagger|JFM\rangle , \end{aligned} \tag{4}$$

where $|JFM\rangle$ and $|J'F'M'\rangle$ are excited states, $|J_0F_0M_0\rangle$ and $|J_1F_1M_1\rangle$ are the initial and final states, respectively, $\hat{e}_1$ and $\hat{e}_2$ are the polarization vectors of exciting and detected light, respectively, $\hat{d}$ is the dipole transition operator, Γ is the natural-decay width of the excited state with energy $E(JFM)$, and $\rho(J_0F_0M_0)$ the densities of the initial states. With nuclear spin $I=0$ and under the assumption of equal population of the ground-state Zeeman sublevels, the following expressions are obtained for the two main transitions involved:

$$i(\tfrac{3}{2},\tfrac{1}{2},\tfrac{1}{2},e_1,e_2) = (1 - \tfrac{1}{2}\sin2\beta'\sin\beta\cos\Omega t)e^{-\Gamma t}, \tag{5}$$

$$\begin{aligned} i(\tfrac{5}{2},\tfrac{3}{2},\tfrac{1}{2},e_1,e_2) \\ = [80 + (3\cos2\gamma'\cos2\beta' + 1)(3\cos2\gamma\cos2\beta + 1) \\ + 3(\cos2\gamma'\cos2\beta' - 1)(\cos2\gamma\cos2\beta - 1)\cos2\Omega t \\ - 60(\sin2\beta'\sin2\beta + \tfrac{4}{5}\cos2\beta'\cos2\beta\sin2\gamma'\sin2\gamma) \\ \times \cos\Omega t]e^{-\Gamma t}, \end{aligned} \tag{6}$$

where $\Omega = (\beta/\hbar)gH$, β is the Bohr magneton, $\hbar$ is Planck's constant divided by 2π, and β, β' is the angle between the fast axis of a quarter-wave plate and the axis of a linear polarizer defining polarizer and analyzer. β, β' defines the ratio between the minor and the major axis of the ellipse as $\tan\beta = b/a$. γ, γ' defines the direction of the major axis relative to the quantization axis. In case of pure circularly-polarized light, $\beta = \beta' = \frac{1}{4}(n+1)\pi$, $n = 0, \pm 1$. In case of π light, $\gamma = \psi - \frac{1}{2}\pi$, with $\beta = \beta' = n\pi$, $n = 0, \pm 1$, and $\psi(\psi')$ is the angle between the ion beam and the orientation of the axis of a linear polarizer. The quantization axis is along the external magnetic field, which, again, is perpendicular to the ion beam. Here it is assumed that the Zeeman splitting is linear, i.e., $E = \beta gMH$, an assumption easily justified, since the fine-structure splitting is of the order of 2000 cm^{-1} and since the applied field is rather weak.

EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in Fig. 2. The 60-keV isotope separator at the University of Aarhus was used to produce a beam of 50-keV $^{138}Ba^+$ ions. The ion beam was mechanically collimated, using two square slits placed symmetrically around the beam focus. The current was measured in a Faraday cup. Typical currents obtained in a 3×3-mm^2 beam were 1–2 μA. An optical system consisting of a lens, a linear polarizer, and a $\frac{1}{4}\lambda$ plate followed by a McPherson model 218 monochromator equipped with a Peltier-cooled EMI 6256S photomultiplier tube detected the light emitted perpendicular to the ion beam. Two pairs of Helmholtz coils produced a magnetic field perpendicular to both the direction of observation and the ion beam. The large Helmholtz coils made possible a dc offset of 40 G, whereas the smaller set of coils, which was controlled by

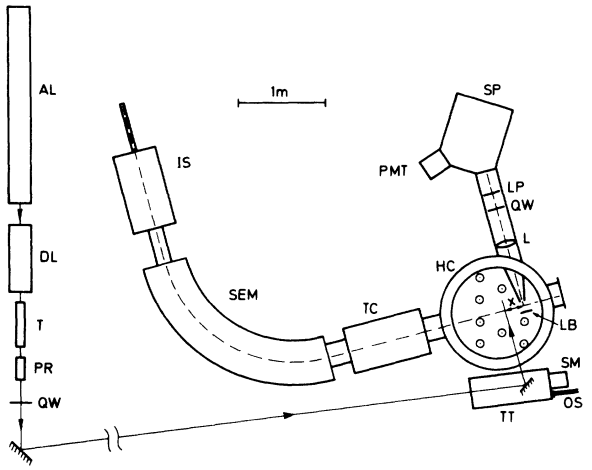

FIG. 2. Experimental setup: AL, argon-ion laser; DL, dye laser; T, telescope; PR, polarization rotator; QW, quarter-wave plate; TT, translation table; OS, distance controller; SM, step motor; LB, light baffle; HC, Helmholtz coils; L, lens; LP, linear polarizer; SP, spectrometer; PMT, photomultiplier; IS, ion source, SEM, separator magnet; TC, target chamber; and x, distance upstream of the focus of lens.

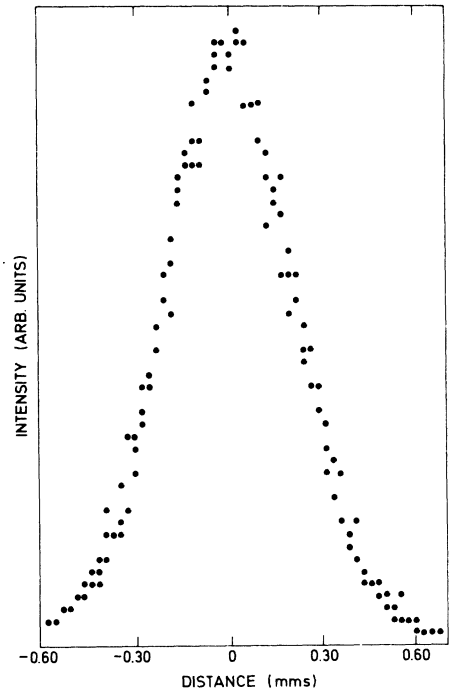

FIG. 3. Experimentally determined laser profile $L(x)$.

a sweep generator, could produce a ramp of up to 90 G. The sweep generator also controlled the x axis of a multiscaler, where several scans could be added. Both of the power supplies used for the Helmholtz coils were current-stabilized. To reduce effects from a change in the coil factor, thermal equilibrium was always ensured before a set of measurements was started. The magnetic field was calibrated via the $4,5p\,^3P_J$ level crossings in He I. A gas cell with a quartz window was placed in the measuring area. The level crossings obtained determined the magnetic field to within 0.2%. H_2 was used as the neutralizing gas, and a 40-keV He^+ beam was used in this calibration.

The Ba^+ ion beam was excited by the output from a cw dye laser which, in turn, was pumped by an argon laser. Rhodamine 6G and Rhodamine B were used as lasing dyes. A polarization rotator and a $\frac{1}{4}\lambda$ plate made possible different types of excitation. During its passage through the ion beam parallel to the detection system, the laser beam could be translated along the beam by means of a translation stage equipped with a step motor. The distance was determined to within 5 μm by means of an electronic distance controller. The laser profile $L(x)$ was determined by translating the laser beam over the focus of the lens through 35-μm slits. Typical laser-profile half-widths were 0.6 mm. A typical laser profile is shown in Fig. 3. The detection profile $D(x)$ was determined by translating a narrow slit through the focus of the lens. Within a g-value measurement, x was kept constant, and the external magnetic field was swept symmetrically around zero. A light-baffle system in front of the lens effectively reduced all background noise. With the laser-output intensities of 20–100 mW used in this study, a background level of 10 counts/sec was typically obtained without the ion beam. With the ion beam typically 2000 counts/sec were obtained when measurements took place approximately three lifetimes after excitation. Light due to rest-gas collisions was not detectable. In order to reduce laser-beam deflections caused by air streaming in the laboratory, it was necessary to enclose the laser beam in a tube system.

DATA REDUCTION

To extract the g values, a trial function,

$$f(i)=\sum_k\sum_J P(J)N(k)\cos\left(q\frac{\beta}{\hbar}G(k)\frac{(x_0+x_1J)(x_2+ix_3)}{v}\right)+x_4+ix_5, \tag{7}$$

was used, where $P(J)$ is the effective normalized experimental profile given by Eq. (2), $q=|M-M'|$, and x_0 is the mean distance between excitation and detection, in millimeters. x_1 is given in mm/step in the experimental profile, J is a summation index running over this profile, x_2 is the initial magnetic field, x_3 the magnetic field step, v the beam velocity, x_4 the linear background, x_5 a slope to take into account the deflection of the ion beam in the external magnetic field, and $G(k)$ the g value with

associated amplitude $N(k)$. A minimum in $\chi^2(N_1, \ldots, N_k, G_1, \ldots, G_k, x_4, x_5)$,

$$\chi^2(N_1, \ldots, N_k, G_1, \ldots, G_k, x_4, x_5) = \sum_i \left(\frac{f_{\text{obs}}(i) - f(i)}{\Delta f_{\text{obs}}(i)} \right)^2 \tag{8}$$

was found where $f_{\text{obs}}(i)$ and $\Delta f_{\text{obs}}(i)$ are the experimental data and error, respectively.

RESULTS AND DISCUSSION

In order to test the alignment of the equipment, the lifetimes of both the $J = \frac{1}{2}$ and $\frac{3}{2}$ levels were measured. The results were in good agreement with previous measurements.[8-10]

As an example of the possibilities of the present technique, the g values of the two fine-structure levels $J = \frac{3}{2}$ and $\frac{1}{2}$ were measured. Results of previously reported measurements, where the classical Zeeman-effect spectroscopy has been used, have been collected by Moore.[14] In Fig. 4 are shown typical results obtained in this work by means of both π and $\sigma^{\pm}$ excitation/detection. The experimental data are least-squares fitted to the trial function of Eq. (7) via the experimentally determined laser and detection profiles. The fit is indicated in Fig. 4 as a solid curve. The g values obtained from this experiment are

$$g(J = \tfrac{3}{2}) = 1.328 \pm 0.008, \quad g(J = \tfrac{1}{2}) = 0.672 \pm 0.006.$$

These values are in excellent agreement with those previously published,[14] 1.32 and 0.672, respectively.

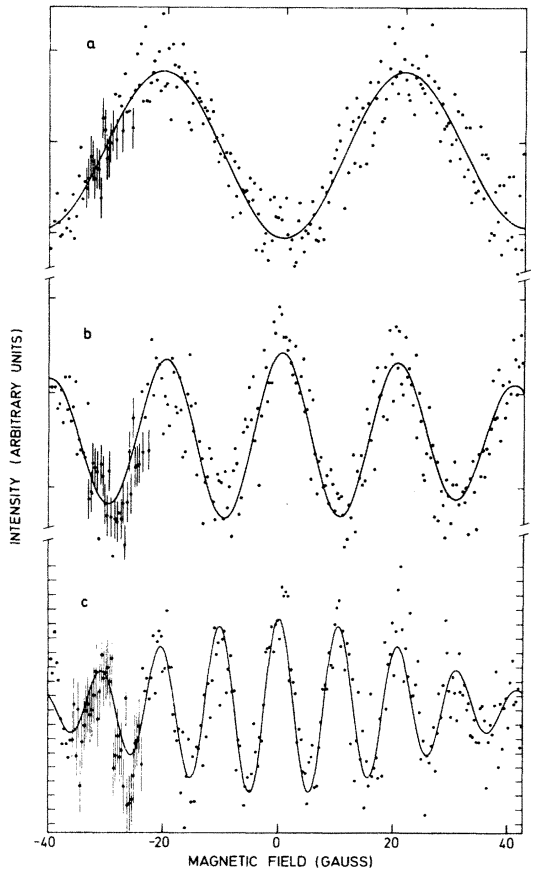

FIG. 4. Examples of the data. Solid curve indicates χ^2 fit, Eq. (8), to the experimental data. (a) Transition $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ in σ^- excitation/detection. Distance upstream of the focus lens is 6.50 mm. (b) Transition $\frac{5}{2} \rightarrow \frac{3}{2} \rightarrow \frac{1}{2}$ in σ^+ excitation/detection. Distance upstream, 7.00 mm. (c) Transition $\frac{5}{2} \rightarrow \frac{3}{2} \rightarrow \frac{1}{2}$ in π excitation/detection. The distance upstream of the focus of the lens is 7.00 mm.

The uncertainties quoted in this work include statistical and systematic errors. The statistical errors typically amount to 0.1–0.2%. The systematic errors included are (i) distance measurement x, measured to within 5 μm, with a drift of ± 10 μm within one measurement (~30 min), (ii) velocity determination of the accelerated barium ions, determined to within 0.1%, (iii) magnetic field calibration using level crossing, to within 0.2%, and (iv) damping, determination of the experiment profile $P(x)$, less than 0.1%. The g values quoted are mean values of up to 20 experiments with different H-field sweeps, distances x upstream of the focus of the lens, and Ba^+ velocities v. The long-term drift of the distance x amounted to ± 30 μm on a total distance of typically 6000 μm over a period of 2 h. All measurements performed were time limited to about 30 min, with determination of the absolute distance x before and after the measurement. In no case was a drift larger than ± 10 μm observed within these small time intervals.

In comparison with a collisional excitation, where the excitation amplitudes are unknown, laser excitation has the great advantage of eliminating the problem of alignment and orientation created in the collisions. If the initial level is isotropically populated, the amplitudes of modulations are given in Eqs. (5) and (6) as a function of excitation and detection parameters. These equations predict that no oscillations with frequency $\Omega = \gamma H$ will be observed when linearly polarized light is responsible for the excitation. In both transitions observed, Ω beats appear in π excitation when the circularly polarized components are observed. This phenomenon was experimentally shown to arise from imperfections in the optical components used. For both the detection and excitation system, the Müller matrix[15] was established and the effective parameters γ, β, γ', and β' determined.

The alignment or orientation is defined as the ratio of peak-to-peak intensity to twice the mean intensity of the oscillations. It was also found that

the imperfections in the optical system, which includes metallic reflections, reduced the amplitudes by about 30%. When these parameters are inserted in Eqs. (5) and (6), good agreement between the theoretically expected amplitudes and experiment is obtained. The alignment produced in the $\frac{3}{2}$ state in π excitation has been found to be about 11%, and the orientation induced by using σ^+ excitation is about 40% for the $\frac{3}{2}$ state and about 30% for the $\frac{1}{2}$ state.

In conclusion, it can be stated that the present technique is a useful extension of the classical level-crossing technique which makes it possible to measure fine and hyperfine structures and g values in a variety of elements at different stages of ionization.

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