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Isotope shift measurements in zirconium by Doppler-free laser polarization spectroscopy*

Ch. Bourauel and W. Rupprecht
Institut für Angewandte Physik der Universität, Wegelerstraße 8, D – 5300 Bonn 1,
Federal Republic of Germany

S. Büttgenbach
Forschungsinstitut der Forschungsgesellschaft für Feingeräte-,
Mikro- und Uhrentechnik e.V., Breitscheidstr. 2b, D-7000 Stuttgart 1,
Federal Republic of Germany

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Isotope shifts in 17 transitions of ZrI have been measured by Doppler-free laser polarization spectroscopy in a hollow cathode discharge. The results were combined with other known isotope shift data of ZrI and tested for consistency in a multidimensional King plot. By including values of $\delta\langle r^2\rangle$ deduced from optical isotope shifts mass and field shifts were separated, and improved values of $\delta\langle r^2\rangle$ could be determined from the King plot. The results for the isotope pairs A, A′ are as follows; 90, 92: 0.244(43) fm^2; 92, 94: 0.176(20) fm^2; 94, 96: 0.126(23) fm^2.

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1. Introduction

In this paper we report on the isotope shifts of 17 transitions in ZrI connecting states of the even configurations $4d^2\,5s^2$ and $4d^3\,5s$ and the odd configurations $4d^3\,5p$ and $4d^2\,5s\,5p$. These laserspectroscopic measurements in the frequency range of the laser dye Rhodamine 110 are the first part of a detailed investigation of the isotope shift (IS) and the hyperfine structure (hfs) of the neutral Zirconium atom.

While the IS of ZrI has been studied for many transitions [1–5] up to now, hfs measurements have been performed only for the low lying even-parity states $a^3F_{2,3,4}$ and $a^5F_{2,3,4,5}$ [6] and the odd-parity states y^5G_5 [7] and z^3F_2 [4]. In order to improve values of the mean square nuclear charge radii which had been determined from conventional optical spectroscopy [8] we performed a multidimensional King plot [9] including the results of this work as well as those reported in [2, 3, 5]. The measurements of Heilig et al. [2] and of Aufmuth and Haunert [3] were performed by means of a photo-electric recording Fabry-Perot spectrometer using enriched isotope samples. Applying intermodulated optogalvanic laserspectroscopy [10] in a hollow cathode discharge Chevalier and Gagné [5] were able to resolve the four even isotopes of natural abundant Zirconium with a linewidth of about 20 MHz. However, as Zr shows relatively low sputtering yields [11] it was difficult for them to observe the hfs of ^{91}Zr$(I=5/2)$ by this technique.

The present measurements were performed by laser polarization spectroscopy [12] in a hollow cathode discharge since this technique had been found to be superior with respect to the signal-to-noise ratio compared to intermodulated fluorescence and intermodulated optogalvanic spectroscopy [13].

2. Experimental apparatus

A schematic diagram of the experimental set-up is shown in Fig. 1. The light of a single mode ring dye laser (Coherent Radiation 699-21) pumped by an argon ion laser is sent through a hollow cathode dis-

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* Dedicated to Professor Siegfried Penselin on the occasion of his 60th birthday

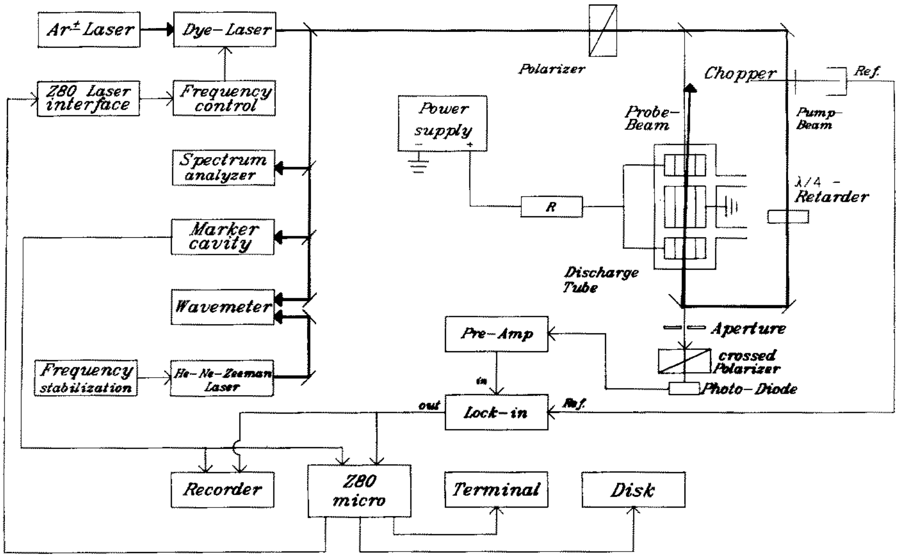

Fig. 1. Experimental set-up for Doppler-free laser polarization spectroscopy in a hollow cathode discharge

charge. The hollow cathode used for our experiments is similar to that designed by Guthöhrlein and coworkers [14, 15]. It consists of a cathode cylinder made of pure aluminium, in which a Zr tube with an inner diameter of 5 mm was inserted, and two symmetrically positioned anodes. In order to reduce the discharge noise as well as the observed linewidth of the polarization signals the hollow cathode was cooled by liquid nitrogen.

The optical arrangement differs only slightly from the one used by Wieman and Hänsch [12]. After passing a Glan-Thompson polarizer the linearly polarized laser beam is divided into two counterpropagating beams which cross the hollow cathode discharge. The stronger pump beam is circularly polarized employing a quarter wave plate, while the weaker probe beam reaches the photodetector after passing a second Glan-Thompson polarizer which is nearly crossed with respect to the first one. The pump beam is mechanically chopped at 3.7 kHz.

The resulting polarization signal was detected by a photodiode, preamplified and then fed to a lock-in amplifier. There was no need of spectral filters. Fluorescent light from the hollow cathode discharge and back-scattered light from the cell windows were blocked by an aperture. Birefringence of the cell windows due to internal stress was reduced by squeezing them with adjustable clamps [12]. By doing this we reached an extinction ratio of the Glan-Thompson polarizers of about 10^4.

The dye laser performance was monitored with a 2 GHz FSR scanning Fabry-Pérot interferometer, and a digital wavelength meter was used to measure the laser frequency with respect to a Zeeman stabilized He–Ne laser. The dye laser sweep was monitored with a 60 MHz FSR nonconfocal reference cavity [16]. The whole experiment is controlled by a Z 80 microcomputer system [17], which scans the laser frequency over the desired frequency range and records the polarization spectra and the calibration fringes of the reference cavity simultaneously.

3. Measurements and results

Figure 2 shows the polarization spectrum of the transition $4d^2\,5s^2\,a^3\,P_2 - 4d^2\,5s\,5p\,y^1\,D_2$ ($\lambda = 538.514$ nm) together with the transmission peaks of the non-confocal marker cavity. The lines of the four even isotopes are resolved with good signal-to-noise ratio. In addition, some of the hfs components of the odd isotope ^{91}Zr are visible. The hollow cathode discharge was operated with a Krypton pressure of 0.6 mbar, a current of 200 mA, which was necessary to achieve sufficient sputtering of Zr atoms, and a voltage of about 700 V. The spectrum was recorded with a laser intensity of 0.5 W/cm^2; the beam had a cross section of approximately 0.1 cm^2. The observed linewidth was 33 MHz (FWHM), the lineshapes are pure Lorentzian.

In general, seven spectra were recorded for each transition. The mean values of the isotope shifts of the 17 investigated lines are compiled in Table 1. The shifts are given with respect to the isotope ^{90}Zr. The quoted errors for the isotope shifts are statistical errors determined from all the spectra taken for each transition and they correspond to three standard deviations.

Our measurements comprise the wavelength region between $\lambda = 536$ nm and $\lambda = 553$ nm. There is

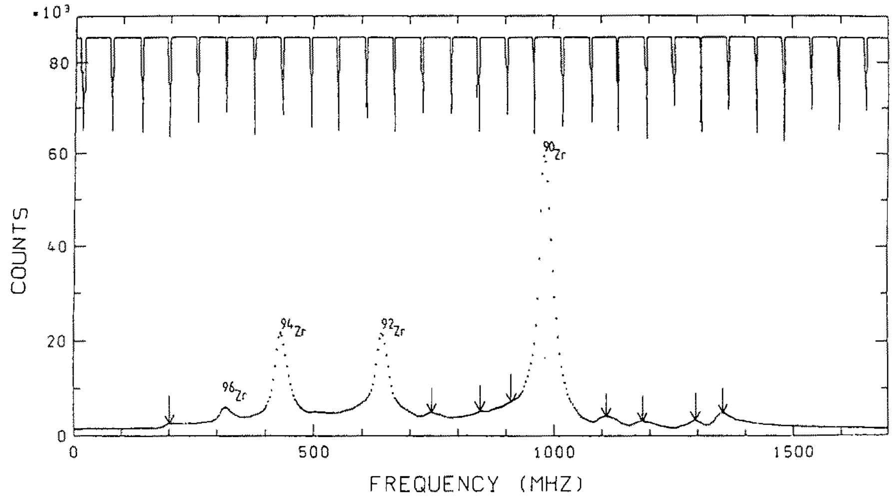

Fig. 2. Polarization spectrum of the transition $a^3P_2-y^1D_2$ at 538.514 nm. Hfs components of ^{91}Zr are marked with an arrow

Table 1. Experimental isotope shifts of Zirconium. A 3σ error is quoted for all transitions. A negative sign indicates that the lighter isotope is shifted towards higher frequencies

Transition	Wavelength	Isotope shifts [MHz]		
	[nm]	90–92	90–94	90–96
$4d^2\,5s^2\;a^3F_4-4d^2\,5s\,5p\;\;z^5D_3$	543.776	−272.4 (3.0)	−432.9 (3.6)	−512.4 (6.3)
$4d^2\,5s^2\;a^3P_1-4d^2\,5s\,5p\;\;y^3D_1$	536.256	−360.8 (3.6)	−590.7 (3.0)	−721.9 (3.6)
$4d^2\,5s^2\;a^3P_2-4d^2\,5s\,5p\;\;y^1D_2$	538.514	−351.1 (4.5)	−570.3 (5.1)	−687.8 (3.6)
$4d^3\,5s\;\;a^5F_1-4d^2\,5s\,5p\;\;y^5D_0$	547.740	−152.3 (5.1)	−328.7 (6.3)	−520.9 (8.4)
$4d^3\,5s\;\;a^5F_1-4d^2\,5s\,5p\;\;y^5D_1$	544.041	−153.7 (3.6)	−331.0 (4.5)	−518.8 (5.7)
$4d^3\,5s\;\;a^5F_1-4d^2\,5s\,5p\;\;y^5D_2$	536.939	−158.7 (7.5)	−337.6 (3.3)	−524.0 (12.0)
$4d^3\,5s\;\;a^5F_2-4d^2\,5s\,5p\;\;y^5D_1$	548.609	−158.3 (9.0)	−338.2 (13.5)	−533.0 (17.4)
$4d^3\,5s\;\;a^5F_2-4d^3\,5p\;\;y^3F_2$	538.237	−183.7 (8.1)	−375.8 (10.5)	−564.0 (17.1)
$4d^3\,5s\;\;a^5F_2-4d^3\,5p\;\;y^3F_3$	539.118	−164.8 (2.4)	−346.8 (3.9)	−541.5 (4.5)
$4d^3\,5s\;\;a^5F_3-4d^2\,5s\,5p\;\;y^3D_2$	553.230	−198.1 (2.4)	−396.5 (4.8)	−593.7 (6.3)
$4d^3\,5s\;\;a^5F_3-4d^2\,5s\,5p\;\;y^5D_2$	548.083	−161.8 (3.6)	−342.4 (4.2)	−533.4 (5.7)
$4d^3\,5s\;\;a^5F_4-4d^2\,5s\,5p\;\;y^3D_3$	551.711	−172.4 (12.0)	−386.1 (12.0)	−573.4 (12.0)
$4d^3\,5s\;\;a^5F_4-4d^2\,5s\,5p\;\;y^5D_3$	544.857	−166.1 (9.3)	−356.3 (12.3)	−548.0 (13.8)
$4d^3\,5s\;\;a^5F_5-4d^2\,5s\,5p\;\;y^5D_4$	540.762	−160.9 (4.8)	−340.2 (2.4)	−534.3 (3.0)
[ref. 2]		−159.0 (9.0)	−345.0 (9.0)	−549.0 (18.0)
$4d^3\,5s\;\;a^5F_5-4d^3\,5p\;\;y^3F_4$	551.805	−165.9 (10.2)	−342.0 (8.1)	−521.3 (17.4)
$4d^2\,5s^2\;a^1D_2-4d^2\,5s\,5p\;\;y^3D_3$	538.665	−316.3 (5.7)	−522.2 (6.3)	−649.1 (5.7)
$4d^2\,5s^2\;a^1D_2-4d^3\,5p\;\;y^3F_2$	540.513	−326.1 (4.5)	−533.4 (5.4)	−651.7 (9.0)

only one transition in this region which has also been investigated by other authors. The results for this transition are in good agreement (see Table 1).

In Table 2 values of the changes in mean square nuclear charge radii deduced by different authors are compared. Heilig and Steudel [8] and Aufmuth and Haunert [3] determined $\beta\delta\langle r^2\rangle$ from optical IS data by applying the Goudsmit-Fermi-Segrè formula. Using the theoretical value $\beta=1.23$ of the screening constant [3] one finds the $\delta\langle r^2\rangle$ values given in Table 2.

We determined improved values of $\delta\langle r^2\rangle$ in the following way. The IS data from this work as well as those from [2, 3, 5] were combined and tested for consistency in a multi-dimensional King plot. This fit procedure, which has been described by Palmer et al. [9], is a generalization of the well known two-dimensional King plot. The input data for the King plot computer program MULTIKING[1] consisted of

Table 2. Comparison of $\delta\langle r^2\rangle$ obtained by several authors

$\delta\langle r^2\rangle$ [fm²]			Reference
90–92	92–94	94–96	
0.246 (49)	0.174 (43)	0.130 (37)	[3]
0.246 (49)	0.170 (46)	0.133 (42)	[8]
0.244 (43)	0.176 (20)	0.126 (23)	This work
0.223 (40)	0.276 (50)	0.559 (100)	[19]

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[1] This program was kindly supplied by C.W.P. Palmer

the mass factors $M(A)\,M(A')/[M(A)-M(A')]$, which were calculated by using the atomic masses M(A) given in the atomic mass table of Wapstra and Bos [18], and the IS data of the 50 transitions included in the fit.

In order to obtain consistency in the fit between the measurements of different authors, the errors were chosen as follows. For the data of Aufmuth and Haunert [3] and for the data of this work we used one third of the quoted 3σ error, while for the data of Heilig et al. [2] we used the quoted 3σ error. For the data of Chevalier and Gagné we used the errors quoted by these authors.

As "reference transition" we chose the values of $\beta\delta\langle r^2\rangle$ from [8]. This resulted in a separation of mass and field shifts. The isotope shifts as well as the $\beta\delta\langle r^2\rangle$ values recalculated in the MULTIKING fit agree very well with the input data, while their error limits could be improved slightly. This result shows not only the overall consistency of the experimental IS data, but also their consistency with the $\beta\delta\langle r^2\rangle$ values deduced by Heilig and Steudel [8]. On the contrary, the values of $\delta\langle r^2\rangle$ determined by Fajardo et al. from elastic electron scattering [19] could not be reproduced in the MULTIKING fit within the experimental error limits.

Further laser polarization spectroscopy measurements in the spectral range of the laser dyes Rhodamine 6 G and Rhodamine 110 are in progress at our laboratory. The aim is a detailed parametric analysis of the IS and hfs of ZrI. For this purpose intermediate coupling wave functions for the $(4d+5s)^4$ model space which take into account effective two- and three-particle configuration interactions have already been developed [20]. On the results of this analysis we shall report in a forthcoming paper.

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