Tensor polarizabilities of the \((nd+ (n+1)s)^3 2D_{3/2,5/2}\) levels in Sc I, Y I, La I and Lu I

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Abstract. A technique based on the detection of nonlinear level-crossing resonances in parallel electric and magnetic fields has been extended to the study of the Stark splittings in the hyperfine levels of the ground multiplets \((nd+ (n+1)s)^3 2D_{3/2,5/2}\) in Sc I, Y I, La I, and Lu I. Values of the tensor polarizabilities of the \(nd\) electrons were evaluated from the experimental results. The variation of the tensor polarizabilities of the \(nd\) electrons in the configurations under study is discussed. Trends of the polarizabilities in different configurations are compared. The ratio between the electric quadrupole constant and the tensor polarizabilities is constant in La and different in Lu, which is caused by a breakdown of the central field model.

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Introduction

This work is a continuation of our programme of a systematic investigation of tensor polarizabilities of ground multiplet levels of free atoms. Previous measurements include determinations of tensor polarizabilities of levels in the ground multiplets of the configurations \(4f^7 5d 6s^2\) in Gd I [1] and \(4f^8 6s^2\) in Sm I [2], in order to test the central field model, and of the configuration \(4f^8 6s^2\) in the rare-earth elements Pr I, Nd I [3], Sm I [2, 4], Tb I, Dy I, Ho I, Er I, and Tm I [3], in order to study the dependence of the polarizability of the \(4f\) electron with an increasing number of \(4f\) electrons. These investigations have revealed tensor polarizabilities of levels of the ground multiplet for all the stable elements of the rare earth atoms with a ground configuration \(4f^8 6s^2\). The values of these tensor polarizabilities show a variation over the \(4f\) shell. On average the tensor polarizability of a \(4f\) electron decreases with an increasing number of \(4f\) electrons [3]. The aim of this paper was to investigate tensor polarizabilities of ground multiplet levels of configurations with one open shell by progressing through a column in the atomic table in order to look for a trend in the single electron tensor polarizability with an increasing main quantum number \(n\).

We now report on the first measurements of the tensor polarizabilities in hyperfine structure levels in the ground multiplets of the configuration \((nd+ (n+1)s)^3\) in Sc I, Y I, La I, and Lu I. Since with the available electric field strengths the Stark splittings of the elements are small in comparison to the hyperfine structure splittings, a Doppler free method was needed for the measurements. The high resolution laser spectroscopic method of nonlinear level-crossing spectroscopy (see, for example, [5]) was used to obtain the values of the tensor polarizabilities of the hyperfine structure levels. The main result of this investigation was that the tensor polarizabilities of the \(nd\) electrons (between \(n = 3\) and 5) were obtained from the tensor polarizabilities of the hyperfine structure levels. This was possible, since only one electron shell with \(l\neq 0\) is open in the configuration under study. The variation of the tensor polarizabilities of the \(nd\) electrons over the quantum number \(n\) is discussed. The influence on the tensor polarizability by built-in completely filled shells of electrons is compared. In addition, the validity of the central field approximation in the ground multiplets was tested and not confirmed for all the investigated elements. The experimental results of the tensor polarizabilities of the \(2D_{3/2}\) levels in Sc I and Y I were confirmed by calculations using experimental oscillator strengths [6, 7].

Measurements

Natural Sc or Y consist only of one stable isotope \(^{45}\text{Sc}\) or \(^{89}\text{Y}\), whereas La or Lu consist of the two stable isotopes \(^{138}\text{La}\) (0.089\%) and \(^{139}\text{La}\) (99.91\%) or \(^{174}\text{Lu}\) (97.41\%) and \(^{176}\text{Lu}\) (2.59\%), respectively. The experimental investigations of the Stark effect were carried out on the strongest isotope of each element, of which the hyperfine structure in the \((nd+ (n+1)s)^3 2D_{3/2,5/2}\) levels of the ground multiplet is known [8–12].
Collinear electric and magnetic fields were used in the experiment. The interaction of homogeneous electric $E$ and magnetic $B$ fields with a free atom which has a nuclear spin $I \neq 0$ leads to an energy shift of the sublevel $|F, m\rangle$. If the Stark shift and the Zeeman splitting are much smaller than the hyperfine splitting, the energy shift $\Delta W(F, m)$ of a sublevel in a collinear electric and magnetic field is given by $[13, 14]$

$$\Delta W(F, m) = g_p \mu_B m B - \{\tilde{\alpha}_0(F) + \tilde{\alpha}_2(F)[3m^2 - F(F + 1)]/[F(2F - 1)]\} \epsilon^2/2,$$

(1)

where $g_p$ is the Landé factor, $\mu_B$ the Bohr magneton, $\tilde{\alpha}_0(F)$ the scalar and $\tilde{\alpha}_2(F)$ the tensor polarizability. In a $(\Delta F = 0, \Delta m = 2)$ or $(\Delta m = 1)$-level-crossing experiment only $\tilde{\alpha}_2$ can be observed, since the term with $\tilde{\alpha}_2$ causes a shift of the sublevels depending on $m^2$. This results in a shift of the magnetic field strength value at which the crossing occurs.

Since the experimental arrangement is similar to the one used in previous experiments $[1, 2]$, only a general outline will be given. In the present experiment an atomic beam of Sc, Y, La, or Lu was produced by electron-bombardment heating of a cylindrical tantalum oven. The atoms in the atomic beam were resonantly excited by the light of a single mode $cw$ dye laser linearly polarized perpendicular or at an angle of 45° to the direction of the magnetic field in order to measure $(\Delta m = 2)$- or $(\Delta m = 1)$-level-crossing signals, respectively. The laser frequency was tuned to one hyperfine structure component of a level of a ground multiplet to any upper level. Both upper and lower states split up in sublevels in the parallel magnetic and electric fields, and crossing between hyperfine Zeeman levels occur in the upper and in the lower states. At values of the field strengths where crossings occur between levels differing by $\pm 1$ or $\pm 2$ in quantum number $m$, the laser light couples one lower with a pair of upper sublevels or one upper with a pair of lower sublevels. The coherence is lost as the degeneracy is removed, and the population of the sublevels changes provided the interaction is nonlinear $[5]$. Contrary to the classical level-crossing effect, crossing in the lower level may also be detected by observing the fluorescence radiation $[15, 16]$.

The fluorescent light was observed as a function of the magnetic field whereas the applied static electric field remained fixed. The magnetic field sweep was controlled by a computer, which also recorded the output signal from the lock-in amplifier. For lock-in detection an amplitude modulation was superimposed on the scanning magnetic field. The modulation depth was chosen so that the observed signal could be related to the derivative of the level-crossing resonance. The signal averaging technique was used to enhance the signal to noise ratio.

One of the experimental difficulties in nonlinear level-crossing spectroscopy is the superposition of the level-crossing signals of the upper and lower levels. In this work only foldover crossings in the lower level were observed, and this was done by choosing suitable transitions. In these the polarizabilities of the excited levels were so large that the level-crossing signals of the sublevels

![Fig. 1. Registration and fit curve of nonlinear level-crossing signals of the lower level in the transition $5d6s^2\ ^2D_{3/2} - 5d^26p\ ^4G_{5/2}$ of $^{139}$La. C: $(\Delta m = 2)$-level-crossing signal in the $^2D_{3/2}$ level between the hyperfine Zeeman levels $|2, \pm 2\rangle, |2, 0\rangle$ at an electric field strength of $\epsilon = 119$ kV/cm](image)

**Table 1. Investigated states and transitions in this work**

<table>
<thead>
<tr>
<th>Element</th>
<th>I</th>
<th>$\lambda$ [Å]</th>
<th>Lower level</th>
<th>$E$ [cm$^{-1}$]</th>
<th>$g_J$</th>
<th>Upper level</th>
<th>$E$ [cm$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Configuration</td>
<td></td>
<td></td>
<td>Configuration</td>
<td></td>
</tr>
<tr>
<td>$^{45}$Sc</td>
<td>7/2</td>
<td>6305.7</td>
<td>$3d4s^2\ ^2D_{5/2}$</td>
<td>168.3$^a$</td>
<td>1.20029$^b$</td>
<td>$3d4s4p\ ^2D_{5/2}$</td>
<td>16022.7$^a$</td>
</tr>
<tr>
<td>$^{89}$Y</td>
<td>1/2</td>
<td>4643.7</td>
<td>$4d5s^2\ ^2D_{3/2}$</td>
<td>0$^+$</td>
<td>0.79929$^c$</td>
<td>$4d5s5p\ ^2F_{3/2}$</td>
<td>21528.6$^e$</td>
</tr>
<tr>
<td>$^{139}$La</td>
<td>7/2</td>
<td>5930.6</td>
<td>$5d6s^2\ ^2D_{5/2}$</td>
<td>0$^+$</td>
<td>0.75755$^d$</td>
<td>$5d^36p\ ^4G_{5/2}$</td>
<td>16856.8$^d$</td>
</tr>
<tr>
<td>$^{175}$Lu</td>
<td>7/2</td>
<td>5736.5</td>
<td>$4f^{14}5d6s^2\ ^2D_{3/2}$</td>
<td>0$^+$</td>
<td>0.79921$^d$</td>
<td>$5d^6s6p\ ^4F_{3/2}$</td>
<td>17427.2$^d$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6055.0</td>
<td>$2D_{3/2}$</td>
<td>1993.9$^d$</td>
<td>1.20040$^d$</td>
<td>$2D_{3/2}$</td>
<td>18504.5$^d$</td>
</tr>
</tbody>
</table>

$^a$ [18]  
$^b$ [8]  
$^c$ [17]  
$^d$ [20]  
$^e$ [19]
Table 2. Tensor polarizabilities of the hyperfine structure levels $\alpha_2(F)$, of the fine structure levels $\alpha_2(J)$, and of the nd electron $\alpha_2(nd)$ (see text). Tensor polarizabilities in kHz/(kV/cm)$^2$

<table>
<thead>
<tr>
<th>Element</th>
<th>Configuration</th>
<th>Crossing $F, m_F, m_{F_2}$</th>
<th>$\alpha_2(F)$</th>
<th>$\alpha_2(J)$</th>
<th>$\alpha_2(nd)$</th>
<th>$\alpha_2(J)_{\text{calc}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{45}$Sc</td>
<td>$3d,4s^2$</td>
<td>$^2D_{3/2}$</td>
<td>$1, \pm 1, 0$</td>
<td>0.15 (1)</td>
<td>1.5 (1)</td>
<td>1.5 (1)</td>
</tr>
<tr>
<td>$^{89}$Y</td>
<td>$4d,5s^2$</td>
<td>$^2D_{3/2}$</td>
<td>$2, \pm 2, 0$</td>
<td>0.55 (5)</td>
<td>0.55 (5)</td>
<td>0.79 (7)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^2D_{3/2}$</td>
<td>$2, \pm 2, \pm 1$</td>
<td>0.72 (7)</td>
<td>0.85 (8)</td>
<td>0.85 (8)</td>
</tr>
<tr>
<td>$^{139}$La</td>
<td>$5d,6s^2$</td>
<td>$^2D_{3/2}$</td>
<td>$2, \pm 2, 0$</td>
<td>0.52 (4)</td>
<td>1.82 (14)</td>
<td>3.2 (2)</td>
</tr>
<tr>
<td>$^{169}$Gd</td>
<td>$4f^7,5d,6s^2$</td>
<td>$^2D_{3/2}$</td>
<td>$1, \pm 1, 0$</td>
<td>0.26 (2)</td>
<td>2.6 (2)</td>
<td>3.1 (2)</td>
</tr>
<tr>
<td>$^{175}$Lu</td>
<td>$4f^{14},5d,6s^2$</td>
<td>$^2D_{3/2}$</td>
<td>$2, \pm 1, 0$</td>
<td>1.00 (8)</td>
<td>3.40 (25)</td>
<td>5.0 (4)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^2D_{3/2}$</td>
<td>$5, \pm 2, \pm 1$</td>
<td>3.31 (25)</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^2D_{3/2}$</td>
<td>$1, \pm 1, 0$</td>
<td>0.54 (5)</td>
<td>5.4 (5)</td>
<td>5.5 (5)</td>
</tr>
</tbody>
</table>

$^a,b$ determined with experimental oscillator strengths of $^a$ [6] and $^b$ [7]

$^c$ [1]

The level-crossing signals were subjected to a least squares fit procedure to extract the exact positions of the level-crossing. The magnetic field positions of the finite field crossings show a linear dependence on the square of the electric field strength (1) within the accuracy of the measurements (see, for example, Fig. 2). Since the $g_J$ factors of the levels of the ground multiplets are known [8, 17, 20] (Table 1), the tensor polarizabilities of the hyperfine structure levels could be derived from the magnetic and electric field values at which the crossings occur. The results are compiled in Table 2. The values represent the means of, typically, ten determinations at different electric field strengths of one or two foldover crossings in a hyperfine level. The quoted errors include the threefold parameter error of the fit procedure as well as systematic errors of the electric (1%) and magnetic (0.1%) field strengths.

Discussion

In this section the variation of the tensor polarizabilities of the $nd$ electrons $\alpha_2(nd)$ in different atoms will be discussed. For this purpose the dependence of the tensor polarizabilities on the nuclear spin I and the total electron spin $S$ have to be eliminated from the measured values $\alpha_2(F)$. These were determined for different hyperfine structure levels in the ground multiplets $(nd+(n+1)s)^3\,^2D_{3/2}, S_{3/2}$ (Table 2). Then the tensor polarizabilities of the $nd$ electrons can be deduced, since only one electron shell with $l=0$ is open in the configuration under study. To do this, first we have derived the tensor polarizabilities of the fine structure levels $\alpha_2(J)$. The tensor polarizabilities of the hyperfine levels $\alpha_2(F)$ can be expressed in terms of the tensor polarizability of a fine structure level $\alpha_2(J)$, as Angel and Sandars have shown.
The values of \( \alpha_2(J) \) are given in Table 2. These are the mean values, if more than one \( \alpha_2(F) \) value was measured in a fine structure level. In Table 2 \( \alpha_2(J) \) values are also indicated as measured in the levels of the ground multiplet \( 4f^75d6s^2\,^9D \) in Gd [1] and two values calculated with experimental oscillator strengths \( f_{s,s,s} \) [6, 7]

\[
\alpha_2(\text{MHz}) = 6.561 \times 10^7 \sqrt{\frac{J(2J-1)(2J+1)}{(2J+3)(J+1)}} \cdot \frac{1}{W(nJ) - W(n'J)} \cdot \sum_{n',J'} (-1)^{J' + J} \cdot \frac{f_{s,s,s}}{(nJ)(n'J')^2} \cdot \langle J' J' \rangle_{12 \ J} \]

where \( W(nJ) \) is the fine structure energy in cm\(^{-1}\). Good agreement is found in a comparison of the calculated values of \( \alpha_2(J) \) with the experimental ones. The signs of the experimental \( \alpha_2(J) \) values were chosen equal to the calculated values.

Provided the ground multiplet in the investigated elements can be understood on the basis of a single configuration model, in the central field model the relation between \( \alpha_2(J) \) and the hyperfine quadrupole coupling constant \( B(J) \) [2]

\[
B(J)/\alpha_2(J) = -\langle nll|C^2|nll\rangle \cdot 2e^2 Q \cdot \langle r^{-3}\rangle /\alpha_2(nll) \]  

should then be a constant for all the levels of a configuration with only one open shell. Any deviation points to a breakdown of the central field model. The ratio (4) for Lu and La is given in Table 3. The results do not show satisfactory agreement. This points to considerable perturbations in the \( ^2D \) multiplets. These may be caused by mixing with higher electronic configurations [2]. The single configuration approach is also inconsistent with the observed hyperfine structure in Lu [12], La [11, 22], Y [9] and Sc [23]. Therefore in the following we used the \( (nd+(n+1)s)^3 \) configurations to derive the single electron tensor polarizability \( \alpha_2(nd) \).

Since only one electron shell with \( l=0 \) is open, the relation between the tensor polarizability of a single electron \( \alpha_2(nl) \)

\[
\alpha_2(nl) = -2\epsilon^{-2} \langle nll|H_{\text{ten}}|nll\rangle 
\]

and the tensor polarizability of a fine structure level \( \alpha_2(J) \) of the configuration \( (nd+(n+1)s)^3 \)

\[
\alpha_2(J) = -2\epsilon^{-2} \left( \frac{J}{-J} \right)_{2J+1} \cdot \sum_{iL'S'J'} \beta_{iL'S'J'} \cdot \{J\, J \, 2\} \left( J' \, L' \, S' \right) \cdot \langle f_{iL'S'J'} \rangle \cdot U_2 \cdot \langle f_{nL'S'L'} \rangle \cdot \langle nll|H_{\text{ten}}|nll\rangle \]  

is given by

\[
\alpha_2(J) = \alpha_2(nl) \left( \frac{J}{-J} \right)_{2J+1} \cdot \frac{1}{(2J+1)} \cdot \sum_{iL'S'J'} \beta_{iL'S'J'} \cdot \{J\, J \, 2\} \left( J' \, L' \, S' \right) \cdot \langle f_{iL'S'J'} \rangle \cdot U_2 \cdot \langle f_{nL'S'L'} \rangle \cdot \langle nll|H_{\text{ten}}|nll\rangle \]  

\( H_{\text{ten}} \) is the Stark operator of the tensor polarizability. The reduced matrix elements of \( U^2 \) have been tabulated by Nielsen and Koster [24], and \( \beta_{iL'S'J'} \) are the mixing coefficients given by [25–28]. The results are shown in Table 2. The values of the two levels of the ground multiplet show good agreement for Y, La and Lu. The results of the levels of the ground multiplet in Gd are similar, except for the value of the \( J=5 \) level (Table 2). The tensor polarizability of this level is caused only by the small admixture of the \( 4f^75d6s^2\,^9D5 \) level in the \( 4f^75d6s^2\,^9D5 \) level. For a detailed discussion of the results in Gd the reader is referred to [1].

Apart from our systematic measurements of the tensor polarizability \( \alpha_2(nd) \) for \( nd \) electrons with \( n \) between 3 and 5 of a ground configuration, up to now only the tensor polarizability of the ground configurations \( npns^2 \) has been measured systematically for some elements. In this series it was found that the absolute values of the tensor polarizability of the \( npns^2 \) configurations of Al [29], Ga [30], In [31], and TI [32] increase with increasing \( n \) (Table 4). This finding was confirmed by calculations with (3) using experimental oscillator strengths [33–36] (Table 4). In contrast, the tensor polarizability \( \alpha_2(nd) \) shows unexpected variation over \( n \) (Table 2). Especially the \( \alpha_2(4d) \) value of Y deviates from the expected trend of increasing \( \alpha_2(nd) \) values with increasing \( n \), since the value of Y is smaller than the value of Sc. A look at the II spectra of Sc, Y, and La shows that the ground configurations of these spectra are \( 3d4s \) (Sc), \( 5s^2 \) (Y), and \( 5d^2 \) (La). This confirms the very different binding of the \( nd \) electrons in the investigated elements. Since in the investigated levels only the \( nd \) electron determines

| Table 3. Values of the ratio \( B(J)/\alpha_2(J) \) of La and Lu for all levels of the ground multiplet \( (nd+(n+1)s)^3 \) \( ^2D_{3/2,5/2} \) (see text) |
|-----------------|-----------------|-----------------|
| Element        | Level           | \( B(J) \) [MHz] | \( \frac{B(J)}{\alpha_2(J)} \) [MV/cm^2] |
| \( ^{139}\text{La} \) | \( ^2D_{3/2} \) | 44.781 (14) \(^a\) | 0.024 (2) |
|                 | \( ^2D_{5/2} \) | 54.213 (13) \(^a\) | 0.021 (2) |
| \( ^{173}\text{Lu} \) | \( ^2D_{3/2} \) | 1511.39865 (69) \(^b\) | 0.44 (3) |
|                 | \( ^2D_{5/2} \) | 1860.66161 (92) \(^b\) | 0.35 (3) |

\(^a\) [11]  
\(^b\) [12]
the value of the tensor polarizability, one cannot expect a systematic trend in the \( \alpha_2(nd) \) values with \( n \). In contrast to this, the ground configurations of Al II, Ga II, In II, and Tl II are \( n s^2 \), and therefore the systematic trend is not surprising.

The tensor polarizability of the 5d electron was derived for the elements La, Gd [1], and Lu (Table 2). The difference in the configuration of the three elements is without a 4f electron, with a half-filled 4f shell, and with a completely filled 4f shell. Although the 4f electrons in the configurations \( 4f^7(\text{6}S) \) \( 5d \) 6s 2 and \( 4f^{14}(\text{1}S_0) \) \( 5d6s^2 \) have no direct influence on the tensor polarizability by reason of symmetry, we see a change in the polarizability due to screening effects. The smaller value of the tensor polarizability of Gd than the value of La can be explained by a stronger binding of the 5d electron in Gd, which is caused by the imperfect screening of the nuclear charge by the 4f electrons [37]. In Lu the binding of the 5d electron is still smaller than in La, and this leads to the greater value of the tensor polarizability in Lu. That the built-in of a filled 4f shell increases the tensor polarizability of a single electron was also found in the value \( \alpha_2(6p) \) derived from the excited levels 6s 6p \( 1P \) in Ba [38] and \( 4f^{14}6s6p \) \( 1P \) in Yb [39]. The absolute value of Yb (\( \alpha_2(6p) = -14.3(1.4) \) kHz/(kV/cm)²) is greater than the absolute value of Ba (\( \alpha_2(6p) = -10.79(29) \) kHz/(kV/cm)²). In contrast to this finding, one can see that the built-in of a filled \( nd \) shell decreases the absolute value of the tensor polarizability of a single electron. This is shown by a comparison of the tensor polarizabilities of the \( np \) electrons derived from the \( 3P \) levels of the \( 5d \) 6s 2 of the configurations \( 4s4p \) \( (\text{Ca}, \alpha_2(4p) = -6.4(1.7) \) kHz/(kV/cm)² [40]) and \( 3d^{10}4s4p \) \( (\text{Zn}, \alpha_2(4p) = -3.6(2) \) kHz/(kV/cm)² [21]), \( 5s5p \) \( (\text{Sr}, \alpha_2 = -12.2(1.6) \) kHz/(kV/cm)² [40]), and \( 4d^{10}5s5p \) \( (\text{Cd}, \alpha_2 = -3.5(2) \) kHz/(kV/cm)² [21]), or \( 6s6p \) \( (\text{Yb}, \alpha_2 = -12.0(4) \) kHz/(kV/cm)² [41]) and \( 5d^{10}6s6p \) \( (\text{Hg}, \alpha_2 = -3.16(4) \) kHz/(kV/cm)² [42]).

All the effects of a completely filled shell of \( nd \) electrons or 4f electrons on the tensor polarizability of a single electron can be explained by the difference in the radial wave functions, which arises because of specific features of the effective potential for 4f electrons. This consists of two wells separated by a potential barrier [43, 44]. The outer well is broad and shallow. The inner well is much narrower and deepens and widens with increasing nuclear charge. For \( Z > 57 \), bound states in the inner well appear, and they cause the 4f electron wave functions to be drawn from the outer into the inner well (lanthanide contraction) [44, 45]. No comparable effects occur for the wave functions of \( s, p, \) or \( d \) electrons. Consequently, the 4f electrons in the lanthanides are strongly bound at radii appreciably smaller than those of the \( n = 5 \) and \( n = 6s \) and \( p \) electrons.

**Conclusion**

The nonlinear level-crossing method was used to measure the tensor polarizabilities in the hyperfine levels of the ground multiplet \( (nd + (n + 1)s)^3 \) \( 2D_{3/2,5/2} \) in Sc I, Y I, La I, and Lu I. The tensor polarizability \( \alpha_2(nd) \) of Sc, Y, and La shows no uniform trend. The variation of \( \alpha_2(nd) \) in La, Gd, and Lu is given by screening effects of the 4f electrons. The built-in of a completely filled 4f shell increases the tensor polarizability of a single electron, whereas the built-in of a completely filled shell of \( nd \) electrons decreases the absolute value of the tensor polarizability.

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