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Hyperfine splitting and isotope shift in the optical transition $4f^7 6s^2\ ^8S_{7/2} \rightarrow 4f^7 6s6p\ ^6P_{5/2}$ of ^{151,153,155}Eu isotopes and electromagnetic moments of ^{155}Eu

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Abstract. The laser-induced resonance fluorescence in an atomic beam was used in order to measure the hyperfine splitting of the $4f^7 6s^2\ ^8S_{7/2}$ and $4f^7 6s6p\ ^6P_{5/2}$ levels in ^{151,153,155}Eu isotopes. The hfs constants A and B of the unstable ^{155}Eu were determined for the first time: $A_{\rm exp}(155,\ ^8S_{7/2}) = -8.78(3)$ MHz, $A_{\rm exp}(155,\ ^6P_{5/2}) = -261.68(11)$ MHz and $B_{\rm exp}(155,\ ^6P_{5/2}) = -951(1)$ MHz. With these data and after corrections for second-order hyperfine structure perturbations the nuclear moments of ^{155}Eu were deduced: $\mu(155) = 1.520(2)$ n.m. and $Q_{\rm s}(155) = 2.494(20)$ b. In addition new and more precise values of the hfs constants of the excited state for the stable ^{151,153}Eu were obtained. They are as follows: $A_{\rm exp}(151,\ ^6P_{5/2}) = -591.92(9)$ MHz, $B_{\rm exp}(151,\ ^6P_{5/2}) = -354.58(97)$ MHz and $A_{\rm exp}(153,\ ^6P_{5/2}) = -263.19(8)$ MHz, $B_{\rm exp}(153,\ ^6P_{5/2}) = -922.07(79)$ MHz. The hyperfine anomalies $^{151}\Delta^{153} = 0.74(4)\%$ and $^{153}\Delta^{155} = 0.27(34)\%$ were extracted from the corrected hfs constants.

PACS. 21.10.Ky Electromagnetic moments – 31.30.Gs Hyperfine interaction and isotope effects, John-Teller effect

1 Introduction

The contemporary laser spectroscopic methods with their high resolution and high sensitivity solve the problem how to access to the basic nuclear ground- and isomeric states in an effective and unique way. Some of the important nuclear properties (*e.g. ms* charge radii changes or multipole nuclear moments) of many elements and in long isotopic chains have been extracted by measurements of the isotope shifts (IS) and hyperfine splitting (hfs) in the optical spectra of atoms and ions. The study of the magnetic dipole and electric quadrupole coupling constants has, in the last years, been a subject of special interest: they provide interesting information about the shape and magnetic moment, as well as about the spatial distribution of the electric currents in the nucleus.

Europium is an element which has many radioactive isotopes with long lifetime. There are 11 various isotopes between ^{145}Eu and ^{155}Eu with $T_{1/2}$ ranging from a few days to a few years. From a spectroscopic point of view, this makes them candidates for systematic hfs investigations in a long isotopic chain. The ^{155}Eu is of particular interest since it is known to have a strongly deformed nucleus, as observed by collinear laser-ion beam spectroscopy [1]. The experiment described below is a first one concerning the hfs of the atomic ^{155}Eu and is devoted to a more precise measurement of the nuclear moments of this isotope.

Hyperfine splitting measurements in the $4f^7 6s^2\ ^8S_{7/2}$ ground and in the $4f^7 6s6p\ ^6P_{5/2}$ excited states are reported. The second-order corrections to the experimental values of the hfs have been calculated. The corrected values of the magnetic-dipole, $A_{\rm corr}$, and electric-quadrupole, $B_{\rm corr}$, constants have been determined and the nuclear moments μ and $Q_{\rm s}$ of ^{155}Eu have been deduced.

Some additional results on the investigated optical transition have been also obtained and are reported here:

(i) a non vanishing hyperfine anomaly between ^{151}Eu and ^{153}Eu and a tendency for an existence of hf anomaly between ^{153}Eu and ^{155}Eu in the $^6P_{5/2}$ level;
(ii) new and more precise values of the hfs constants of the excited state for the stable ^{151}Eu and ^{153}Eu;
(iii) new values of the isotope shifts and *ms* charge radii changes for the isotope pairs ^{151,153}Eu and ^{153,155}Eu.

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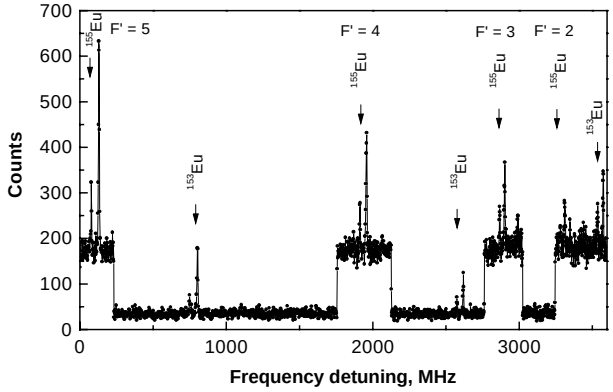

Fig. 1. The transition at 564.58 nm of ^{155}Eu and ^{153}Eu. The scan velocity for recording the ^{155}Eu peaks was about ten times lower than the one of the ^{153}Eu peaks in order to obtain higher statistics. The flag-pattern groups of the ^{155}Eu are indicated by the total quantum number F' of the upper level.

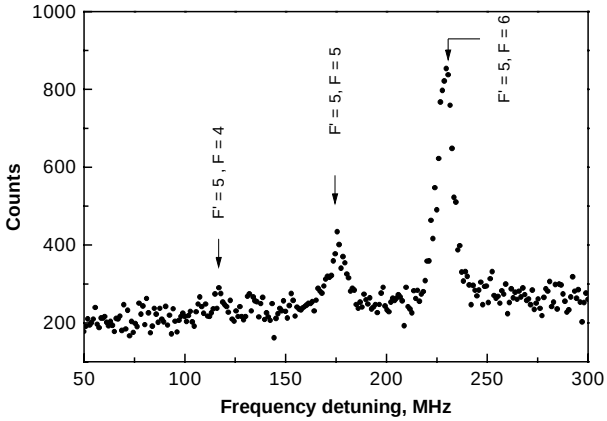

Fig. 2. The most intensive peak group $F' = 5 \rightarrow F = 6, 5, 4$ of the hfs spectrum of ^{155}Eu at $\lambda = 564.58$ nm recorded with higher resolution.

2 Experimental method

An off-line experimental method was used, based on the detection of the laser-excited fluorescence in a well collimated atomic beam. A detailed description of the experimental set-up is given elsewhere [2, 3]. A CW dye laser pumped with an Ar ion laser was operated using R6G dye. To reduce the Doppler broadening, both the atomic and the laser beams were collimated and the laser beam crossed the atomic beam perpendicularly. The laser-induced resonance fluorescence was detected with a photomultiplier, operating in a single photon counting mode, and recorded in a computer. For accurate determination of peak positions a confocal Fabry-Perot interferometer with a free spectral range of 150 MHz producing a set of frequency markers recorded simultaneously with the fluorescence spectra was used.

The ^{155}Eu ($T_{1/2} = 4.76$ years) isotope was obtained in the reaction

$$^{154}\mathrm{Sm}(n,\gamma)^{155}\mathrm{Sm} \xrightarrow{\beta^-} {}^{155}\mathrm{Eu}.$$

A drop of $Eu(NO_3)_3$ water solution containing 10^{14} atoms of ^{155}Eu was dried on a tantalum foil. The heating of the sample in a Ta crucible to a temperature of about 1800 °C resulted in a thermal dissociation of the $Eu(NO_3)_3$ and provided a stable beam of ^{155}Eu for about 45 min. Care was taken to minimize the abundance of the stable ^{153}Eu isotope, the hf structure of which, as was expected, slightly overlapped the one of the ^{155}Eu. The ratio of ^{153}Eu to ^{155}Eu in the sample was about 10:1. In order to perform reliable measurements with the radioactive ^{155}Eu the apparatus was tested with samples containing only the stable ^{153}Eu in quantities down to 10^{14} atoms. In this way all the experimental conditions were optimized.

The optical transition from the Eu ground state $4f^7 6s^2\ {}^8\mathrm{S}_{7/2}$ to the $4f^7 6s6p\ {}^6\mathrm{P}_{5/2}$ with $\lambda = 564.58$ nm was studied. The large difference in the hfs of both levels of the investigated transition results in a typical flag-pattern spectrum (Fig. 1). The narrow triplet groups are defined by the small hfs of the ground $^8\mathrm{S}_{7/2}$ state [4] (see Fig. 2). The laser frequency was tuned over a region of about 5 GHz to cover the whole ^{155}Eu hf structure and one scan took typically 20 min. An experimental FWHM of the detected resonance lines, composed by the natural and laser line widths and the Doppler broadening, was about 8–9 MHz. Under this condition all hf structure components were completely separated.

Table 1. Experimental, ΔW_{exp}, values of the hf splitting of the lower $^8\mathrm{S}_{7/2}$ and the upper $^6\mathrm{P}_{5/2}$ levels for ^{155}Eu. Here $\delta = \Delta W_{\mathrm{cal}} - \Delta W_{\mathrm{exp}}$. All energies are in MHz.

$4f^7 6s^2\ {}^8\mathrm{S}_{7/2}$				$4f^7 6s6p\ {}^6\mathrm{P}_{5/2}$			
F	F'	ΔW_{exp}	δ	F	F'	ΔW_{exp}	δ
5	6	53.61(21)	0.03	4	5	1879.07(71)	−0.05
4	5	44.27(47)	−0.29	3	4	989.44(66)	0.23
3	4	33.89(99)	0.81	2	3	443.26(87)	−0.57
2	3	27.33(218)	−1.57	1	2	153.12(285)	0.65
1	2	17.00(184)	0.04	0	1	33.10(63)	0.34

3 Data analysis and results

The procedure of the experimental spectra analysis was described in details in our previous paper [5]. The computer processing of the linearized experimental spectrum allowed for accurate determination of the frequency intervals between the various peaks. The precision was lower only for some peak positions from the last two groups of the spectrum, because of their low intensity. The line shape was assumed to be a Voigt-profil; 8 different spectra, recorded on 4 samples have been used in the analysis.

The experimental values of the hfs spacing for both ground $4f^7 6s^2\ {}^8\mathrm{S}_{7/2}$ and excited $4f^7 6s6p\ {}^6\mathrm{P}_{5/2}$ states of ^{155}Eu are presented in Table 1. The errors quoted correspond to one standard deviation of the spread between different spectra. The systematic error due to the uncertainty of the free spectral range of the frequency calibration interferometer is negligible small. From these values

Table 2. Hfs constants for the lower and upper levels of the optical transition with $\lambda = 564.58$ nm. The table gives experimental $A_{\rm exp}$, $B_{\rm exp}$ and corrected $A_{\rm corr}$, $B_{\rm corr}$ hfs constants (all in MHz). $^{(*)}$ denotes our adopted value for the electric quadrupole constant of the ground $^8S_{7/2}$ level for ^{155}Eu. For comparison, for the stable Eu isotopes the most precise known values are presented, too.

	$A_{\rm exp}$	$A_{\rm corr}$	$B_{\rm exp}$	$B_{\rm corr}$	Ref.
$^8S_{7/2}$					
^{151}Eu	−20.0523(2)		−0.7012(12)		[4]
^{153}Eu	−8.8532(2)		−1.7852(35)		[4]
^{155}Eu	−8.78(3)	−8.78(3)	−1.861(20)$^{(*)}$	−1.877(36)	this work
$^6P_{5/2}$					
^{151}Eu	−591.92(9)	−591.16(9)	−354.58(97)	−348.38(97)	this work
	−590.7(5)	−591.0(5)	−354(4)	−350(4)	[7]
^{153}Eu	−263.19(8)	−262.94(8)	−922.07(79)	−921.80(79)	this work
	−263.3(3)	−263.2(3)	−919(3)	−920(3)	[7]
^{155}Eu	−261.68(11)	−261.48(11)	−951(1)	−950.4(1.0)	this work

the magnetic dipole, $A_{\rm exp}$, and electric quadrupole, $B_{\rm exp}$, constants for both levels of the transition were evaluated in a least-squares fitting (LSF) procedure using the expression [6]:

$$W(F) = \frac{1}{2}AC + B\frac{3C(C+1)/4 - J(J+1)I(I+1)}{2J(2J-1)I(2I-1)} \quad (1)$$

where $C = F(F+1) - I(I+1) - J(J+1)$ and J, I and F are the quantum numbers of the electronic angular momentum, nuclear spin and the total atomic angular momentum, respectively. The experimental values $A_{\rm exp}$ and $B_{\rm exp}$ are listed in Table 2. The quality of the fit was checked by the normalized $\chi = (\chi^2)^{1/2}$ which in different cases was in the range of 0.55–1.04. The differences between the experimental and recalculated values of the hf spacings for the ground $^8S_{7/2}$ and the excited $^6P_{5/2}$ levels are within the experimental errors (see Tab. 1).

It should be noted that in the case of ^{155}Eu the hf coupling constants of the ground state were not obtained by the usual two-parameters LSF procedure according equation (1). The experimental method used in the present work does not meet the standard of the more precise double rf-laser spectroscopy and in the case of small hfs gives relative bad accuracy. However, the precision of the magnetic dipole constant is of a special interest. To obtain more accurate value for A we used the fact that (i) the ratio B'/B of the electric quadrupole constants for both levels of an optical transition should be equal for different isotopes of a given element, and (ii) the contribution of the quadrupole interaction to the total hfs in the investigated case is very small. Thus, the value of $B(155,\ ^8S_{7/2})$ was first found using the relation:

$$\frac{B(151,\ ^8S_{7/2})}{B(151,\ ^6P_{5/2})} = \frac{B(153,\ ^8S_{7/2})}{B(153,\ ^6P_{5/2})} = \frac{B(155,\ ^8S_{7/2})}{B(155,\ ^6P_{5/2})} \quad (2)$$

With $B(151)$, $B(153)$ for the ground $^8S_{7/2}$ state from [4] and $B(151)$, $B(153)$, $B(155)$ for the excited $^6P_{5/2}$ state from the present work (Tab. 2) it follows for $B(155,\ ^8S_{7/2})$: −1.881(6) MHz and −1.841(4) MHz from the first and second ratio respectively. The disagreement of about 2% between these $B(155,\ ^8S_{7/2})$-values does not influence the value of the magnetic dipole constant $A(155,\ ^8S_{7/2})$ extracted then by an one-parameter LSF to the experimental hfs intervals given in Table 1. This is because the hf splitting of the $^8S_{7/2}$ level is determined predominantly by the magnetic dipole hf interaction. Thus, one can fix the $B(155,\ ^8S_{7/2})$ as a mean value from the both obtained values and estimate the uncertainty of the adopted $B(155,\ ^8S_{7/2})$ to be 0.020 MHz so that both experimental $B(155,\ ^8S_{7/2})$-values are covered (see Tab. 2).

From the hfs constants obtained in the present work the values of nuclear moments of ^{155}Eu can be estimated using the known magnetic and electric quadrupole moments of ^{153}Eu: $\mu = 1.5330(8)$ n.m. and $Q_{\rm s} = 2.412(21)$ b [8,9]. For the electric quadrupole moment of ^{155}Eu we obtain $Q_{\rm s}(155) = 2.495(19)$ b as a weighted average of values obtained with the $B(155)$ value of the ground, as well as of the excited state.

The situation with the magnetic dipole moment is in general different. As known [10], the ratio of the hf coupling constant A for two isotopes is not exactly equal to the ratio of their gyromagnetic factors $g_I = \mu/I$ as a result of the finite extension of the nucleus and the distribution of nuclear magnetization. The effect is expressed in the hf anomaly $^1\Delta^2$ between two isotopes 1 and 2 which is generally small, at most a few percent:

$$^1\Delta^2 = 1 - \frac{A(1)/A(2)}{g_I(1)/g_I(2)}\,. \quad (3)$$

Because $s_{1/2}$ and $p_{1/2}$ electrons are the only ones with non-zero densities near the nucleus, they are the only ones that will be affected by the finite nuclear size [10]. Therefore, one could expect that the $^1\Delta^2$ is nearly zero for the EuI $^8S_{7/2}$ ground state with its paired s-electrons and for the investigated Eu isotopes with equal spins, $I = 5/2$, equation (3) can be expressed in the form:

$$^1\Delta^2 = 1 - \frac{A(1,\ ^6P_{5/2})/A(2,\ ^6P_{5/2})}{A(1,\ ^8S_{7/2})/A(2,\ ^8S_{7/2})}\,. \quad (4)$$

Thus, we estimate the magnetic moment of ^{155}Eu from the ratio of the magnetic dipole coupling constants for

Table 3. Ratios of the magnetic dipole coupling constants of the investigated levels for 151, 153 and 155 europium isotopes and the hf anomaly of the excited state according to equation (4). $A(151)$ and $A(153)$ values for the ground state have been taken from [4].

	$^8S_{7/2}$	$^6P_{5/2}$	$^1\Delta^2$, %	
$A(151)/A(153)$ corr	2.26498(8)	2.2483(8)	0.74(4)	this work
		2.245(3)	0.88(13)	[7]
$A(153)/A(155)$ corr	1.0083(34)	1.0056(5)	0.27(34)	this work

Table 4. Experimental IS and field shift (FS) for the optical line with $\lambda = 564.58$ nm and the *ms* charge radii changes for the investigated Eu isotope pairs.

A	A'	IS, MHz	FS, MHz	$\delta\langle r^2\rangle$, fm^2	Ref.
151	153	−3662.0(9)	−3687.2(12.6)	0.563(2)	this work
				0.602	[17]
				0.542	[19]
153	155	−674.7(10)	−699.3(12.3)	0.108(2)	this work
				0.075	[17]
				0.109	[19]

the ground state (see Tab. 3): $\mu(155) = 1.520(2)$ n.m. Our experimental results are in a good agreement with the first estimation of $\mu(155)$ and $Q_s(155)$ [1], they are, however, more precise than the earlier ones.

In the course of our test experiments the hfs of the $^6P_{5/2}$ excited level for ^{151}Eu and ^{153}Eu was also measured and the results are summarized in Table 2. The accuracy of the experimental A and B values was improved in comparison with the previous paper [7,11] and it is sufficient for reliable information on the hf anomaly $^{151}\Delta^{153}$ to be obtained.

However, from earlier investigations of the hfs of Eu isotopes [7, 12–14] it is known that the mixing of electronic states with different J quantum numbers may play an important role and usually cannot be neglected in the interpretation of hfs measurements. To take this into account the "repulsion effect" among hfs sublevels with the same quantum number F, belonging to different levels, has been calculated using second-order perturbation theory.

In order to correct the hfs constants for the J off-diagonal hfs perturbations we have used the procedure described in [12–14]. The $4f^7 6s6p\ ^6P_{5/2}$ level exhibits only a slight amount of configuration interaction. This simplified greatly our hfs calculations for this levels. As the shift of the $4f^7 6s6p\ ^6P_{5/2}$ sublevels is mainly determined by the "repulsion" of the neighbouring $4f^7 6s6p\ ^6P_{3/2,7/2}$, $^{10,8}P_{7/2,9/2}$ levels, the eigenvector compositions of these levels were analysed [15]. The non-linear corrections cannot be implemented in a linear fitting procedure used for the linear expansion of hfs interactions. Therefore, the energy shifts δW_F were first used to correct the measured hfs intervals. Then the hfs constants A_{corr} and B_{corr} were calculated by the fitting to the corrected hfs intervals. These values are presented in Table 2 with uncertainties including only the experimental statistical errors.

We would like to emphasize that the second-order corrections to hfs intervals for the $^8S_{7/2}$ level are very small (less than 1 kHz) and have been neglected. This is due to two reasons: (i) an electronic configuration with paired s-electrons which in general can lead only to a vanishing hf anomaly (nearly 0.003(19)% in the case of EuI ground state for the ^{151,153}Eu isotope pair) and (ii) the absence of other atomic levels in the near vicinity of the ground state which can mix the electronic states *via* the hf interaction. The hfs constants A_{corr} and B_{corr} showed in Table 2 for the ground state of ^{155}Eu presents just our recalculation of these constants with using B_{corr} values of ^{151,153,155}Eu for the exited state (see Eq. (2)). The B_{corr} value of the ground state changed for about 0.85, but it does not influent on the value of constant A, *e.g.* after the fitting with a new fixed constant B_{corr} we obtained the same value of constant A. This indicates that the second order corrections does not influent on the value of ^{155}Eu magnetic moment. Moreover, our recalculation of $Q_s(155)$ value with constants B_{corr} for exited and ground states gives the same weighted average value of $Q_s(155) = 2.494(20)$ b as it was obtained with uncorrected B_{exp}.

Table 3 shows the hf anomalies $^{151}\Delta^{153}$ and $^{153}\Delta^{155}$ of the excited state of the investigated transition relative to the ground state according to equation (4). It is seen that the hf anomaly in the $^6P_{5/2}$ level is relatively large for the isotope pair ^{151,153}Eu. This is in agreement with the experimental results of other authors (see *e.g.* [7]) but does not correspond to the recent theoretical predictions of [16]. As about the hf anomaly for the isotope pair ^{153,155}Eu, it is in the limits of the errors, because of the relatively large experimental uncertainties of the magnetic dipole constant for the ground state of ^{155}Eu.

With the hfs constants from Table 2 using the measured intervals between hfs components of ^{151}Eu and ^{153}Eu, as well as of ^{153}Eu and ^{155}Eu the determination of the corresponding isotope shifts (IS) $\delta\nu^{A,A'} = \nu(A) - \nu(A')$ is straightforward. The ms charge radii changes were also deduced under the assumption that the investigated optical transition is a pure $s^2 \rightarrow sp$-transition [11] and with the following parameters: $F(Z)_{s^2-sp} = -6.55$ GHz/fm^2 from [17] and specific mass shift $SM = (0 \pm 0.5)$ NM (NM – normal mass shift) as suggested in [18]. The results are presented in Table 4. We emphasize (i) the very good agreement with the *ms* charge radii changes in reference [19], predicted on the basis of a combined analysis of two types of experimental data on nuclear charge radii: radii measured from electron scattering and muonic atom spectra and radii changes determined from optical IS, and (ii) the more precise experimental values of $\delta\langle r^2\rangle$, than those presented in the review paper [17].

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