# Ground-state hyperfine-structure measurements of unstable $Eu^+$ isotopes in a Paul ion trap

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Hyperfine separations in unstable $Eu^+$ ions of mass 148, 149, and 150 have been measured in laser-microwave double-resonance experiments in a Paul ion trap. In spite of the small available quantities of the isotopes, the experimental uncertainties are of the order of $10^{-8}$ or below, which is of the same order as in earlier measurements on stable isotopes of $Eu^+$. Extensive second-order perturbation calculation is required to obtain coupling constants for magnetic-dipole ($A$) and electric-quadrupole ($B$) interactions. The uncertainties are a few times $10^{-7}$ for $A$ and $10^{-3}$ for $B$. The experiments are a step in an attempt to determine the differential hyperfine anomaly (Bohr-Weisskopf effect) in a long chain of isotopes. [S1050-2947(97)02807-2]

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## I. INTRODUCTION

In the central-field approximation the electronic wave functions depend on only one property of the nucleus: its total charge $Z$. Thus two isotopes of the same element should have the same electronic wave functions and the matrix elements of hyperfine structure (hfs) magnetic dipole operator should be identical for both isotopes. In such a case the relationship

$$\frac{A(1)}{A(2)}=\frac{\mu_I(1)I(2)}{\mu_I(2)I(1)}=\frac{g_I(1)}{g_I(2)} \tag{1}$$

should hold [1], where $A(1)$, $\mu_I(1)$, $I(1)$ and $g(1)$ are the magnetic dipole hfs interaction constant, nuclear magnetic-dipole moment, nuclear spin, and nuclear $g$ factor, respectively, of isotope 1 (and the same for isotope 2). All the quantities appearing in Eq. (1) can be measured. When these measured values are applied, Eq. (1) is often found to be incorrect because the distribution of magnetization over the extended nuclear volume has not been taken into account. Deviations from the above simple relation are expressed in terms of a hyperfine anomaly $^1\Delta^2$:

$$^1\Delta^2=\frac{A(1)g_I(2)}{A(2)g_I(1)}-1. \tag{2}$$

Nuclear models could in principle predict the magnitude of $^1\Delta^2$. Fujita and Arima [2] have performed calculations considering the magnetic interaction energy as a product of the nucleon current and the vector potential of the electrons at the nucleus. The agreement of calculated values of $^1\Delta^2$ to experimental values, however, is not satisfactory. Surprisingly good agreement for the case of Hg, which is the only case where experimental data on several isotopes are available, is obtained by a simple empirical rule of Moscowitz and Lombardi [3], that the difference between the calculated and measured value of $A$ is inversely proportional to the nuclear magnetic moment. Quite generally, however, the status of our understanding of the hyperfine anomaly is rather poor. Büttgenbach [4] has reviewed the status of experiment and theory some time ago, and to our knowledge no significant progress has been made on the experimental as well as on the theoretical side since then. This may be caused by the fact that reliable data exist only for the stable isotopes of an element. A renewed interest in the subject may arise when systematic and precise data on a long chain of isotopes are available. This has motivated us to start spectroscopic experiments on a chain of isotopes.

In order to determine the hyperfine anomaly $^1\Delta^2$, the ratios $A(1)/A(2)$ and $g_I(2)/g_I(1)$ have to be measured by two independent experimental methods. The experiment of laser-microwave double resonance in a Paul trap allows one to measure hfs $A$ constants with relative uncertainties of $10^{-10}$ or below [5]. $g$-factor measurements using Penning traps and strong superimposed magnetic fields have resulted in values accurate to about $10^{-6}-10^{-7}$ [5]. This is more than adequate for the investigation of hyperfine anomalies since the typical magnitude of $^1\Delta^2$ is $10^{-2}-10^{-3}$.

In our investigations we have chosen Eu as a candidate since it offers many isotopes of long lifetime and nonzero nuclear spin (see Table I). The unstable isotopes are produced by nuclear reactions at the CERN-ISOLDE facility at rates of $10^8-10^{10}$ $s^{-1}$ after mass separation. After collection on platinum or rhenium filaments there is sufficient time to perform spectroscopic experiments off the production area at our laboratory at the University of Mainz. The Eu nuclei ($Z=63$) can be described in a frame of a single-particle model since it has a one-proton hole in the $5d$ subshell. At $N=82$ we have a closed neutron shell. Data on isotope shifts are available from collinear laser spectroscopy [6], which gives data on deformation parameters of the unstable nuclei.

Recently we have demonstrated that the hyperfine structure of radioactive ions can be investigated successfully using the ion trapping technique [7]. In our previous work on the stable isotopes $^{151}Eu^+$ and $^{153}Eu^+$ we have shown that

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TABLE I. List of long-living Eu isotopes.

| Isotope | Nuclear spin | Half-life |
|---|---|---|
| $^{145}$Eu | $\frac{5}{2}$ | 5 d |
| $^{146}$Eu | 4 | 4.6 d |
| $^{147}$Eu | $\frac{5}{2}$ | 24 d |
| $^{148}$Eu | 5 | 54 d |
| $^{149}$Eu | $\frac{5}{2}$ | 93 d |
| $^{150}$Eu | 5 | 36 *a* |
| $^{151}$Eu | $\frac{5}{2}$ | stable |
| $^{152}$Eu | 3 | 13 *a* |
| $^{153}$Eu | $\frac{5}{2}$ | stable |
| $^{154}$Eu | 3 | 8.8 *a* |
| $^{155}$Eu | $\frac{5}{2}$ | 5 *a* |
| $^{156}$Eu | 0 | 15.2 d |
| $^{157}$Eu | $\frac{5}{2}$ | 15 h |
| $^{158}$Eu | 1 | 0.77 h |
| $^{159}$Eu | $\frac{5}{2}$ | 0.31 h |

magnetic-dipole hfs constants $A$ for the ground state can be obtained by the laser-microwave double-resonance technique with an accuracy of about $10^{-8}$. Using the ratio $g_I(2)/g_I(1)$ measured directly by Evans, Sandars, and Woodgate [8], we were able to determine the value of hyperfine anomaly with a high accuracy: $^{151}\Delta^{153} = -0.006\,63(18)$ [9].

In this work we report the results of laser-microwave double-resonance experiments in a Paul trap as an important step towards the determination of hyperfine anomalies. It allowed us to give the ratio $A(1)/A(2)$ for several isotopes of Eu. The magnetic-dipole constants $A$ have been corrected for the effect of mixing of the electronic wave functions via the nucleus using the method described in [9]. As a by-product we obtain accurate electric-quadrupole constants $B$ for the isotopes $^{148}$Eu$^+$, $^{149}$Eu$^+$, and $^{150}$Eu$^+$.

## II. EXPERIMENTAL SETUP

The experimental setup is nearly identical to that in Ref. [9] with some improvements in the optical detection system. We used a hyperbolic-shaped Paul trap of 4 cm diameter, driven by a (nominal) 400-kHz oscillator. Isotopes were collected in quantities of $10^{12}$ particles each on a platinum or rhenium filament at the ISOLDE-CERN facility. The filament was placed in a slot in the lower end cap of the trap. Heating of the filament for typically 10 s to temperatures of about 1200 °C produced ions by means of surface ionization. An estimated fraction of $10^{-6}$ of them was slowed down by collisions with neutral buffer gas molecules ($N_2$) at $10^{-4}$ mbar and confined for several hours in the trap. The buffer gas collisions served also for moderate collisional cooling of the ions and for quenching of long-living metastable electronic states into which laser-excited ions may decay.

A laser of typically 2 mW power from a frequency-doubled Ti:sapphire laser passes the trap in the $x$-$y$ plane through two holes of 4 mm diameter and excites the ion cloud. Laser-induced fluorescence is collected through a mesh end-cap trap electrode, a collection optic, a pinhole for stray-light suppression, and a GaAs photomultiplier tube. The output of the photomultiplier is transferred for further handling to a personal computer, which also provided the timing sequence for microwave frequency steps. Typical counting rates at resonant excitation were several $10^3$ Hz from a cloud of about $10^5$ ions.

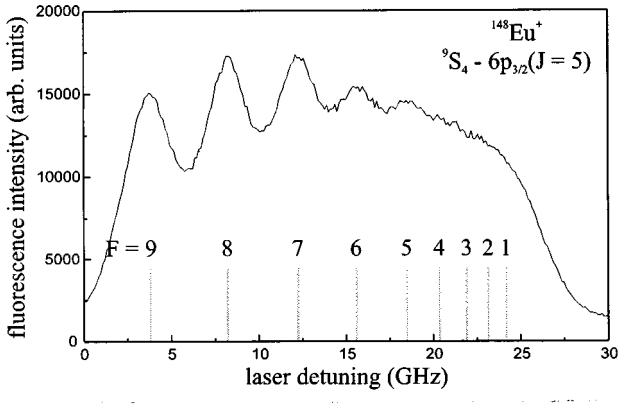


FIG. 1. Doppler-limited laser-induced fluorescence spectrum of trapped radioactive $^{148}$Eu$^+$ ions.

## III. MEASUREMENTS

A measurement cycle contained the following steps. After storage the light from the laser was tuned across the $^9S_4$-$^9P_5$ electric-dipole resonance at 382 nm. The fluorescence spectra showed partially resolved hyperfine splittings of this line. Figure 1 shows the isotope $^{148}$Eu$^+$ as an example. The spectral resolution is limited by Doppler broadening. From the linewidth of 5 GHz (full width at half maximum) a temperature of 3800 K is derived. The resolution was sufficient to excite selectively several hyperfine components of the ground state and deplete them by optical pumping. Microwave-induced $\Delta F = \pm 1$ transitions from neighboring hyperfine levels are detected by the increase in fluorescence intensity. For high microwave power we observe three resonances according to the different polarizations of the microwaves (Fig. 2). For high-resolution measurements we concentrated on the $\pi$ component. At lower

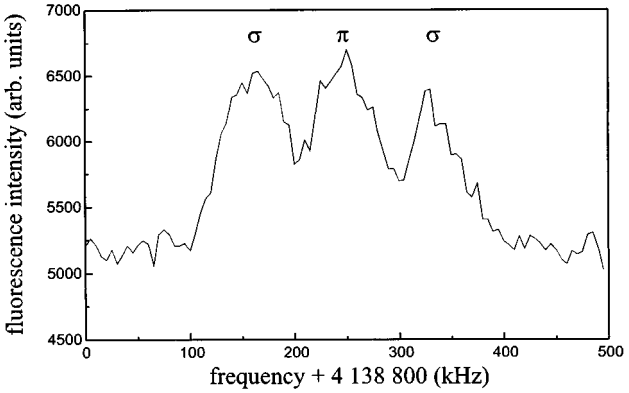


FIG. 2. Low-resolution scan of the $F=8 \rightarrow F=7$ microwave-induced hyperfine transition in the $^9S_4$ ground state of radioactive $^{148}$Eu$^+$, showing the $\Delta_{mF}=0,\pm 1$ components in a residual magnetic field of about 6 $\mu$T.

microwave power the Zeeman substates in a residual $B$ field of typically 6 $\mu$T were resolved (Fig. 3). Finally, the central part of the Zeeman pattern was recorded with high resolution and the center of the resonance pattern was determined. The linewidth was limited to a few 100 Hz by power broadening and by magnetic-field inhomogeneities.

A typical time for the measurement of one hyperfine splitting was about 5 h including repetitive scans to improve the signal-to-noise ratio. We have measured for each isotope five different hyperfine intervals. As a consequence, we had more data than needed to determine the hyperfine coupling constants. The additional data served, however, for consistency checks for the corrections from the complex perturbation calculation. Table II lists the experimentally obtained splittings between different hyperfine levels. The error bars are purely statistical ($1\sigma$) and result from the average of several measurements. The numbers are corrected for the influence of a quadratic Zeeman shift. Only in a few cases the correction exceeded slightly the statistical uncertainty.

We note that in spite of the low number of available particles the experimental uncertainties are generally smaller than those obtained in our previous experiment on the stable isotopes of $Eu^+$ [9]. This is due to technical improvements in the apparatus, in particular in the optical detection system.

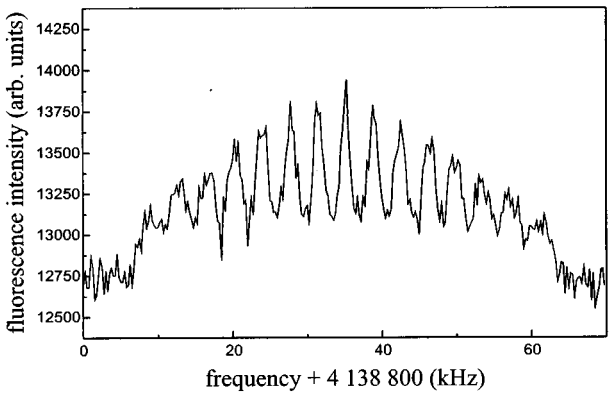


FIG. 3. High-resolution microwave scan of the $\Delta m_F=0$ part of the $F=8\rightarrow F=7$ hyperfine transition in the ground state of $^{148}Eu^+$ in a residual field of about 6 $\mu$T.

## IV. DETERMINATION OF THE hfs CONSTANTS

From our earlier investigations of the hyperfine structure in the ground state of europium isotopes $^{151,153}Eu^+$ as well as $^{150}Eu^+$ [7,9] it is known that hfs interactions that mix electronic states with different $J$ quantum numbers play an important role and cannot be neglected. This manifests itself in deviations from the Landé interval rule. To take this into account the ''repulsion effect'' between different hfs sublevels with the same quantum number $F$ has been calculated using second-order perturbation theory. In this case the energy shift is given by

$$\delta W_F(\Psi,J)=|\langle\Psi,JIFm_F|H_{\rm hfs}|\Psi',J'IFm_F\rangle|^2/[E(\Psi,JF)-E(\Psi',J'F)], \quad (3)$$

where $|\Psi\rangle$ denotes the real fine structure $SLJ$ state (wave function in intermediate coupling) written in $SL$ basis. $I,F,M$ are good quantum numbers characterizing the real hfs state. The Hamiltonian aperator $H_{\rm hfs}$ can be written as

$$H_{\rm hfs}=\sum T_N^K \sum_{\kappa,k} (T_{\rm el}^{(\kappa k)K}+X^{(\kappa k)K}), \quad (4)$$

where $T_N^K$ is the nuclear multipole moment operator of order $K$ [10], $T_{\rm el}^{(\kappa k)K}$ is the electronic one-body hfs operator [10], $X^{(\kappa k)K}$ is the two-body hfs operator, and $\kappa$ and $k$ are the tensor rank in spin and orbit space, respectively (for more details see [11]). The matrix element in the numerator of Eq. (3) can be expressed as a function of one- and two-body hfs radial parameters $a_{\rm nl}^{\kappa k}$, $b_{\rm nl}^{\kappa k}$, $a_i$, and $b_i$, respectively [11]. To

TABLE II. Experimentally obtained transition frequencies between ground-state hyperfine transitions in different $Eu^+$ isotopes

| Isotope | Transition $F\rightarrow F'$ | Frequency (Hz) | Uncertainty |
|---|---|---|---|
| $^{150}$Eu | 8→9 | 5 393 263 079(9) | $1.7\times10^{-9}$ |
| | 7→8 | 4 793 088 792(14) | $2.9\times10^{-9}$ |
| | 6→7 | 4 193 240 682(23) | $5.5\times10^{-9}$ |
| | 5→6 | 3 593 677 733(12) | $3.3\times10^{-9}$ |
| | 4→5 | 2 994 358 980(26) | $8.7\times10^{-9}$ |
| $^{149}$Eu | 11/2→13/2 | 10 311 366 164(12) | $1.1\times10^{-9}$ |
| | 9/2→11/2 | 8 721 591 347(8) | $1.0\times10^{-9}$ |
| | 7/2→9/2 | 7 133 524 668(9) | $1.2\times10^{-9}$ |
| | 5/2→7/2 | 5 546 853 094(76) | $1.4\times10^{-8}$ |
| | 3/2→5/2 | 3 961 264 983(91) | $2.3\times10^{-8}$ |
| $^{148}$Eu | 8→9 | 4 657 266 064(9) | $2.0\times10^{-9}$ |
| | 7→8 | 4 139 049 973(8) | $2.0\times10^{-9}$ |
| | 6→7 | 3 621 096 128(15) | $4.2\times10^{-9}$ |
| | 5→6 | 3 103 371 426(43) | $1.4\times10^{-8}$ |
| | 4→5 | 2 585 843 300(13) | $5.1\times10^{-9}$ |

TABLE III. Experimental and corrected values of the hfs constants for the magnetic-dipole ($A$) and electric-quadrupole ($B$) interaction in the ground state $4f^7(^8S)6s\ ^9S_4$ of the unstable isotopes $^{148,149,150}Eu^+$. Columns 3 and 4 correspond to the values of hfs constants uncorrected and corrected for second-order hfs interactions, respectively. Errors given in column 3 are purely statistical. All values are given in hertz.

| Isotope | hfs constant | Uncorrected | Corrected |
|---|---|---|---|
| $^{148}Eu^+$ | $A$ | 517 302 217(5) | 517 281 950(150) |
| | $B$ | 4 579 440(85) | −292 630(1000) |
| $^{149}Eu^+$ | $A$ | 1 585 640 944(10) | 1 585 450 570(250) |
| | $B$ | 9 637 340(240) | −534 850(1900) |
| $^{150}Eu^+$ | $A$ | 599 037 743(12) | 599 010 680(200) |
| | $B$ | 5 698 770(430) | −839 730(3000) |

obtain preliminary values of the magnetic-dipole hfs parameters for each isotope under study we assumed in the first step that

$$\frac{a_{\rm nl}^{\kappa k}(^z\mathrm{Eu}^+)}{a_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)}=\frac{a_i(^z\mathrm{Eu}^+)}{a_i(^{153}\mathrm{Eu}^+)}=\frac{A_{\rm expt}(^z\mathrm{Eu}^+)}{A_{\rm expt}(^{153}\mathrm{Eu}^+)}, \tag{5}$$

where $z=148$, 149, or 150 and $A_{\rm expt}(^z\mathrm{Eu}^+)$ are the experimental values of the hfs magnetic-dipole constant from Table III (column 3). The values of $A_{\rm expt}(^{153}\mathrm{Eu}^+)$, $a_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)$, and $a_i(^{153}\mathrm{Eu}^+)$ were taken from [9]. The procedure described here was applied for each isotope under study. In a second step, the hfs radial parameters $a_{\rm nl}^{\kappa k}$, $b_{\rm nl}^{\kappa k}$, $a_i$, and $b_i$ were used to determine the energy shifts $\delta W_F$ of each hfs sublevel, caused by second-order hfs perturbation. In the calculations, the perturbing states with $J=3$ and 5, like those in [9], were taken into account and wave functions in intermediate coupling were used for the perturbed state as well as for all perturbing states. Improvements in the calculation compared to Ref. [9] were obtained by new and accurate values of the hyperfine splitting of the $^7S_3$ metastable state, which acts as the most important perturbing state. These measurements will be published separately [12].

The nonlinear correction cannot be implemented in a linear fitting routine used for the solution of the linear expansion of the hfs interaction. Therefore, the energy shifts $\delta W_F$ were first used to correct the measured hfs intervals. Then the hfs constants $A_{\rm corr}(^z\mathrm{Eu}^+)$ and $B_{\rm corr}(^z\mathrm{Eu}^+)$ were obtained by fitting to the corrected hfs intervals. This allowed us to calculate the values of the one- and two-body hfs parameters for magnetic-dipole as well as electric-quadrupole hfs interactions using the relations

$$\frac{a_{\rm nl}^{\kappa k}(^z\mathrm{Eu}^+)}{a_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)}=\frac{a_i(^z\mathrm{Eu}^+)}{a_i(^{153}\mathrm{Eu}^+)}=\frac{A_{\rm corr}(^z\mathrm{Eu}^+)}{A_{\rm corr}(^{153}\mathrm{Eu}^+)}, \tag{6}$$

$$\frac{b_{\rm nl}^{\kappa k}(^z\mathrm{Eu}^+)}{b_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)}=\frac{b_i(^z\mathrm{Eu}^+)}{b_i(^{153}\mathrm{Eu}^+)}=\frac{B_{\rm corr}(^z\mathrm{Eu}^+)}{B_{\rm corr}(^{153}\mathrm{Eu}^+)}. \tag{7}$$

TABLE IV. Experimental $A$ ratios $[A(^z\mathrm{Eu}^+)/A(^{153}\mathrm{Eu}^+)]_{\rm corr}$ ($z=148,149,150,151$) for the ground state $4f^7(^8S)6s\ ^9S_4$, necessary for the determination of hyperfine anomalies.

| $z$ | $A_{\rm corr}(^z\mathrm{Eu}^+)/A_{\rm corr}(^{153}\mathrm{Eu}^+)$ |
|---|---|
| 148 | 0.755 634 89(23) |
| 149 | 2.315 993 76(40) |
| 150 | 0.875 022 55(31) |
| 151 | 2.250 034 927(35)[a] |

[a]Reference [9].

The values of $A_{\rm corr}(^{153}\mathrm{Eu}^+)$, $a_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)$, $a_i(^{153}\mathrm{Eu}^+)$, $B_{\rm corr}(^{153}\mathrm{Eu}^+)$, $b_{\rm nl}^{\kappa k}(^{153}\mathrm{Eu}^+)$, and $b_i(^{153}\mathrm{Eu}^+)$ were taken from [9]. The values of the corrected hfs constants are given in Table III (column 4).

## V. DISCUSSION

An analysis of the results presented in Table III and those in Table II in [9] leads to the conclusion that second-order hfs interactions influence the hyperfine structure of the ground state of all unstable isotopes under study in the same way as for the stable isotopes $^{151,153}Eu^+$, despite the difference in nuclear spins: $I(^{150}\mathrm{Eu}^+)=I(^{148}\mathrm{Eu}^+)=5$ and $I(^{149}\mathrm{Eu}^+)=I(^{151}\mathrm{Eu}^+)=I(^{153}\mathrm{Eu}^+)=\frac{5}{2}$. Consideration of the hfs second-order effects did not change strongly the values of magnetic-dipole hfs constant $A$. The differences $\Delta A=[A(^z\mathrm{Eu}^+)/A(^{z+1}\mathrm{Eu}^+)]_{\rm uncorr}-[A(^z\mathrm{Eu}^+)/A(^{z+1}\mathrm{Eu}^+)]_{\rm corr}$ were about $2\times10^{-5}$ only and thus we think that an uncertainty of our calculations cannot influence significantly the precision of corrected $A$ ratios necessary for the determination of hyperfine structure anomalies. The ratios are given in Table IV.

The values of the hfs constant $B$ are particularly sensitive to the second-order hfs corrections [14]. The constants $B$ have changed not only their values but also their sign after the above corrections. The results of our present experiments and calculations allow us to determine the values of nuclear quadrupole moments $Q$ for all investigated isotopes using the conventionally assumed relation between the hfs $B$ constants and quadrupole moments $Q$: $B(^z\mathrm{Eu}^+)/B(^x\mathrm{Eu}^+)=Q(^z\mathrm{Eu})/Q(^x\mathrm{Eu})$ ($x=151$ or 153). As reference we have taken $B(^{151}\mathrm{Eu}^+)=-690\ 612(70)$ Hz or $B(^{153}\mathrm{Eu}^+)=-1\ 759\ 520(180)$ Hz and the nuclear moment values $Q(^{151}\mathrm{Eu})=0.903(10)$ b or $Q(^{153}\mathrm{Eu})=2.412(21)$ b from muonic x-ray measurements [15]. The final results are listed in Table V.

They are in good agreement with the values published by

TABLE V. Spectroscopic nuclear electric quadrupole moments $Q_s$ of the isotopes $^{148,149,150}Eu^+$ (in barn).

| Isotope | $Q_s$ This work | $Q_s$ Other work |
|---|---|---|
| $^{148}Eu^+$ | 0.392(10) | 0.35(6)[a] |
| $^{149}Eu^+$ | 0.716(17) | 0.75(2)[a] |
| | | 0.74[b] |
| $^{150}Eu^+$ | 1.125(27) | 1.13(5)[a] |

[a]Reference [6].
[b]Reference [13].

Ahmad *et al.* In the case of the isotope $^{150}Eu^{+}$ for which the preliminary value $Q(^{150}Eu) = 1.264(23)$ b was reported by us earlier [7], the present additional and more accurate measurements and calculations indicate that the above given $Q$ value should be smaller: $Q(^{150}Eu) = 1.125(27)$ b. This more precise value confirms the result given in [6] (see Table V). On account of the experimental data and accuracy of the hfs second-order corrections we were able to establish the upper limit of the magnetio-octupole interaction constant $C$ and electric-hexadecapole interaction constant $D$ of the ground state of $^{148,149,50}Eu^{+}$ to be not higher than a few hertz.

## VI. CONCLUSION

We have determined the magnetic-dipole ($A$) and electric-quadrupole ($B$) hfs constants in the ground state of the unstable europium isotopes $^{148,149,50}Eu^{+}$. For the magnetic-dipole hfs constants $A$ our results are about two orders of magnitude more accurate than required for the determination of hyperfine structure anomalies at the 1% level.

The present values of electric-quadrupole moments $Q$ obtained from $B$ values are more accurate than those given before [6,7,13]. Moreover, the upper limit of the magnetic-octupole ($C$) and electric-hexadecapole ($D$) interaction constants has been evaluated. Determination of values of the magnetic-octupole and electric-hexadecapole nuclear moments should be possible if optical microwave double-resonance experiments can be performed with the same precision for more levels and the uncertainty of hfs second-order correction calculations can be reduced by more accurate wave functions. Improved wave functions could be obtained in a fine-structure analysis if new fine-structure experimental data are available.

Our experiment demonstrates that precise experimental data on hyperfine-structure separations can be obtained by the ion storage technique even in cases where only very few particles are available and the electronic level structure of the ions is rather complex. We consider our results as an important step towards the systematic investigation of hyperfine-structure anomalies. Complimentary experiments to determine the nuclear $g$ factors for the same isotopes of $Eu^{+}$ using the Penning trap technique are under preparation in our laboratory.

## ACKNOWLEDGMENTS

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