How Accurate Are the "Muonic" Quadrupole Moments in Eu?

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The electric quadrupole hyperfine structure splitting constants B of the Eu isotopes 145–147, 151–153 were measured with collinear laser ion beam spectroscopy in 17 different transitions. From 105 experimental data B values of all levels in Eu II $4f^7(^8S)5d\,^9D_J$, ...$6p(7/2;1/2)_J$, and ...$6p(7/2;3/2)_J$ were determined in enhanced accuracy. The ratio $^{151}B/^{153}B=0.391\,84(22)$ (mean value) significantly deviates from values of muonic measurements but is in good agreement with other atomic ones in Eu I and Eu II. Values for the ratios of quadrupole moments are given.

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In a recent paper published in this journal Sundholm and Olsen [1] have shown that the quadrupole moments Q of the nuclei ^{23}Na and ^{27}Al obtained from atomic measurements do not agree with measurements of muonic x-ray experiments and they "could not avoid the conclusion that the uncertainty of the muonic x-ray transition experiments may be larger than expected."

This statement has far reaching consequences: Many Q moments of radioactive nuclei were determined by optical methods, as these methods are very sensitive and fast. From the splitting factors B of the hyperfine structure (hfs), for example measured with collinear laser ion beam spectroscopy (CLIBS), and the electric field gradient q at the nucleus the values for Q can be derived.

However, the calculation of q is difficult, even if one uses other experimental data as hfs splitting factors A or fine structure splitting factors ζ. So in most cases the following is assumed: (1) q is isotope independent, which means that no hfs anomaly of the B values is present, and thus $^1Q/^2Q={}^1B/^2B$. (2) A reference Q moment $Q_{\rm ref}$ is known from optical measurements on a stable isotope and on an atomic configuration which is best suited for a reliable estimation or an *ab initio* calculation of q. (3) $Q_{\rm ref}$ can be taken from muonic measurements on stable isotopes, as the wave function of the single muon and thus q is almost undisturbed by the other electrons.

The fact that the accuracy of muonic Q values is now under question explains why Sundholm and Olsen used the slightly provocative subtitle, which is used in this paper again.

A disagreement between optical and muonic Q moments may have different causes: (i) inaccurate optical B values; (ii) inaccurate q values due to insufficient treatment of the perturbation of the atomic wave function due to interaction within the atom or due to the quadrupole field of the nucleus (Sternheimer correction [2]); (iii) inaccurate x-ray spectra analysis; (iv) inaccurate estimation of the distortion of the nucleus due to the penetrating muon. One should not forget that the level density in the nucleus is generally higher than the level density of the muonic atom.

In the following we will not try to calculate absolute Q values from B values, as was done in [1], and compare them with muonic Q values. Instead we will compare ratios of optical Q moments with ratios of muonic ones. This has the disadvantage that now the errors of two experimental values enter into the result, but the large advantage is that the difficult and perhaps problematic calculation of q can be avoided.

Best suited for a comparison of muonic and atomic data of quadrupole moments are the stable nuclei ^{151}Eu and ^{153}Eu. They have moderate or large Q moments, so that the relative experimental errors are small: $^{151}Q_\mu=0.903(10)\,e\,$b, $^{153}Q_\mu=2.412(21)\,e\,$b. The muonic values Q_μ are among the most accurate ones ever measured [3].

The two Eu nuclei have a completely different nuclear structure, as ^{153}Eu ($N=90$) is the lightest of the deformed and rigid nuclei, and ^{151}Eu ($N=88$) is the heaviest of the lighter ones which have a similar structure to the magic (radioactive) nucleus ^{145}Eu ($N=82$). (The differences are also visible in the magnetic splitting factors A and the isotope shifts, and thus in the nuclear magnetic moments and the nuclear radii [4,5].) Therefore the chance that an insufficient treatment of the perturbation of the nucleus due to the muon will show up is relatively large.

Recently we have measured hfs spectra of ^{145,146,147,151,152,152m,153}Eu transitions between the two configurations $4f^7(^8S)5d\,^9D$ and $4f^7(^8S)6p_{1/2,3/2}$ in Eu II to obtain data for the hfs anomaly in A factors [6] and second-order effects in J-dependent isotope shifts [7]. As a by-product of the analysis we also got 105 B values, 66 of them for the stable isotopes ^{151,153}Eu. All values were derived from 17 different transitions with completely different hfs spectra and up to 15 resolved hfs components. So the probability of a systematic error due to the evaluation of B from structures with sometimes overlapping components is small. The data are given in Table I for ^{151}Eu and ^{153}Eu.

For the ratios of the B values, which should be equal to the ratios of the Q values, we got consistently the same

TABLE I. B factors of the eleven levels of ^{153}Eu II $4f^7(^8S)5d\,^9D_J$ and $4f^7(^8S)6p$. They were reduced from 105 B factors measured in all 17 transitions and with six isotopes, four of them radioactive. The B factors for the other isotopes can be calculated by multiplication with the ratios given in the last column.

J	$4f^7(^8S)5d\,^9D$	$\ldots 6p_{1/2}(7/2;1/2)$	$\ldots 6p_{3/2}(7/2;3/2)$	A	$^{A}B/^{153}B$
2	275.19(42)		321.85(1.5)	145	0.11684(87)
3	−623.44(43)	305.72(55)	−1272.90(47)	146	−0.0743(20)
4	−549.11(49)	−859.71(79)	394.03(54)	147	0.2185(23)
5	52.22(51)		1196.16(65)	151	0.39184(22)
6	923.99(70)			152	1.18220(49)

values. Its average is $^{151}B/^{153}B=0.39184(22)$. The accuracy of the difference ΔB of the B values of the upper and the lower levels of a transition is higher than that of each of the values alone. The reason is that the B factors are mathematically correlated when an experimental curve is fitted with two A and two B values, the line-shape parameters, and the isotope shift [8]. A slight increase of B of the upper level will shift the calculated hfs components. A corresponding increase of B of the lower level will shift the components partially back to the original position. The correlation between B and other parameters is much smaller. A slight increase of the difference ΔB of the two B values, however, cannot be compensated by any variation of other parameters. Its value is thus well defined; the error is small, smaller than that of each of the two B values.

So we also compared the ratios of the differences ΔB for the two isotopes. The ratio was again consistent and gave

$$^{151}B/^{153}B=0.39191(12)\,.$$

This value is in agreement with the value given above and with values in the literature, obtained by different experimental methods in different configurations of the neutral or the ionized Eu. See Table II.

Finally we tried to find a hfs anomaly of the B values. For the A values it is known that the ratio $^{151}A/^{153}A$ is not exactly that of the magnetic moments and in the investigated levels it is term dependent $\delta(^{151}A/^{153}A) = -1.2\pm0.6\%$ [6,8]. The experimental B values give no obvious hint for a hfs anomaly of the B values.

Sternheimer corrections affect the values for B by more than $\delta B=0.1B$. They are due to polarization of the electron cloud due to the quadrupole moment. Surely this dependency of δB with B cannot go linearly to infinity with increasing B. So we looked for a quadratic correction term in B, which should affect the large, but not the small B values. This kind of term cannot be extracted from our measurements; the value c obtained for this quadratic term cB^2 is $c=(-5.5\pm23)\times10^{-8}$ MHz^{-2}. As the error of c is more than 4 times as large as the value we must conclude that $^{151}B/^{153}B$ is equal to $^{151}Q/^{153}Q$ within the given experimental error limits and thus we cannot remove the discrepancy with the muonic

TABLE II. B-value and Q-value ratios obtained by different experimental methods.

$^{151}Q/^{153}Q$	$^{151}B/^{153}B$	Configuration	Method	Reference
1.2/2.5			Optical hfs	[9]
	0.3928(20)	$4f^76s^2(^8S)_{7/2}$	Atomic beam magnetic resonance	[10]
0.91(15)/2.50(22)	0.368(65)	$4f^76p\,^9P_5$	Optical hfs	[11]
1.16(8)/2.92(20)	0.3973(386)	$4f^76s6p\,^{6,8,10}P$	Optical hfs	[12]
1.12(7)/2.85(18)	0.3930(349)	$4f^76p\,^9P$	Optical hfs	[13]
	0.3922(34)	$4f^75d\,^6P_{7/2}$	Laser atomic beam	[14]
	0.3918(13)	$^8P_{7/2}$	Laser atomic beam	[14]
1.53(5)/3.92(12)	0.3903(175)	$4f^75d6s6p$	Laser atomic beam	[15]
	0.38(4)	$4f^75d\,^9D$	CLIBS	[16]
	0.39002(76)	$Eu^{3+}\,^7F_0$	Optically detected NMR	[17]
	0.39124(77)	$Eu^{3+}\,^5D_0$	Optically detected NMR	[18]
	0.39188(4)[a]	Eu II ^9D mean value	Laser beam radio frequency double resonance	[19]
	0.3914(76)	Eu I ^{10}D mean value	Laser beam radio frequency	
	0.3926(76)	^8D mean value	double resonance	[20]
	0.39184(22)	Eu II mean value	CLIBS	This work
	0.39191(12)		See text	This work

[a]Calculated from B values corrected for higher-order effects, given in [19].

values. We also cannot "avoid the conclusion that the uncertainty of the muonic x-ray transition experiments may be larger than expected."

To complete the picture Table I gives the results of the fit of the 105 experimental B values in respect to the B values of the eleven investigated levels and the six ratios of the Q moments. The values are given for ^{153}Eu. The values for the other isotopes can be calculated with the help of the ratios $^{A}B/^{153}B$. The value $^{151}Q_\mu/^{153}Q_\mu$ $=0.903(10)/2.412(21)=0.3744(53)$ from the muonic measurements [3] is not in accord with the values for the ratios of the quadrupole moments; the ratio of the B values of $^{151}B/^{153}B=0.391\,84(22)$. At least one of the muonic Q moments is not correct by more than three error margins. Therefore we do not give values for quadrupole moments of ^{145,146,147,151,152,153}Eu despite the increased accuracy of the ratios of the Q moments obtained in this work as there is no reliable reference value available.

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