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Hyperfine Anomaly in Eu Isotopes and the Universiability of the Moskowitz–Lombardi Formula

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Abstract: A method for determining the hyperfine anomaly, without using the nuclear magnetic moments, is used on a series of unstable isotopes of Eu. The large number of experimental data in Eu makes it possible to extract the hyperfine anomaly for a number of unstable isotopes. Calculations of the Bohr–Weisskopf effect and hence the hyperfine anomaly are performed using the particle-rotor formalism. The result from the calculations and experiments is compared with other theoretical calculations and the empirical Moskowitz–Lombardi formula. The results show that the Moskowitz–Lombardi formula is not universal.

Keywords: hyperfine structure; hyperfine anomaly; nuclear magnetic dipole moment

1. Introduction

The study of hyperfine structure (hfs) in atoms has provided information of the electromagnetic moments of the nucleus, as well as information on the electronic properties' of atoms [1,2]. The magnetic hfs has in addition proven to give information on the distribution of magnetization in the nucleus through the so called Bohr–Weisskopf effect (BW effect) [3–5]. The influence of the finite size of the nucleus on the hyperfine structure was first considered by Bohr and Weisskopf [3]. They calculated the hyperfine interaction (hfi) of $s_{1/2}$ and $p_{1/2}$ electrons in the field of an extended nucleus and showed that the magnetic dipole hyperfine interaction constant (A) for an extended nucleus is generally smaller than that expected for a point nucleus. The extended charge distribution of the nucleus gives rise to the so-called Breit–Rosenthal effect (BR effect) [6–9]. In this case, the differential BR effect is assumed to be negligible when two isotopes are compared, as expected in the case of light nuclei. Inclusion of the BR effect will not have any influence on the results, since the BW and BR effects show the same behavior. The BR effect is therefore not considered in the following discussion.

Isotopic variations of nuclear magnetic dipole moments become larger than those in the point dipole interaction when there are different contributions to the hfs from the orbital and spin parts of the magnetization in the case of extended nuclei. The fractional difference between the point nucleus hfi constant (A_{point}) and the constant obtained for the extended nuclear magnetization is commonly referred to as the Bohr–Weisskopf (BW) effect [5]. The hfi constant A can therefore be written as:

$$A = A_{point} (1 + \epsilon_{BW}) \quad (1)$$

where ϵ_{BW} is the BW effect and A_{point} is the A constant for a point nucleus. The BW effect is dependent on both nuclear properties, as well as atomic properties, i.e., the electron density within the nucleus. The nuclear part, i.e., the distribution of nuclear magnetization, can be calculated using different nuclear models [4,5]. Because electronic wavefunctions cannot be calculated with high accuracy in complex atoms, as they can be in hydrogen-like ions and muonic atoms, it is not always possible to determine ϵ_{BW} directly. However, it is possible to determine the difference of the BW effect in

two isotopes, the so-called (differential) hyperfine anomaly (hfa). Where one compares the ratio of the measured hfs constants for two isotopes with the independently measured ratio of the nuclear magnetic dipole moments to extract the hfa, ${}^1\Delta^2$, for the isotopes 1 and 2, and a given atomic state:

$$1 + {}^1\Delta^2 = \frac{A^{(1)} \mu_I^{(2)} / I^{(2)}}{A^{(2)} \mu_I^{(1)} / I^{(1)}} \approx 1 + \epsilon_{BW}^{(1)} - \epsilon_{BW}^{(2)} \quad (2)$$

where μ_I is the nuclear magnetic dipole moment and I the nuclear spin. For electrons with a total angular momentum $j > 1/2$, the anomalies may be disregarded as the corresponding wavefunctions vanish at the nucleus. The hfa can show a dependence of the atomic state, a state dependent hfa, where the values for different states can vary significantly. The reason is that the hyperfine interaction consists of three parts [10,11], orbital, spin-orbit, and contact (spin) interaction, where only the contact interaction contributes to the hfa. It is suitable to rewrite the dipole hyperfine interaction constant as:

$$A = A_{nc} + A_c \quad (3)$$

where A_c is the contribution due to the contact interaction of s (and $p_{1/2}$) electrons and A_{nc} is the contribution due to non-contact interactions. The experimental hfa, which is defined with the total magnetic dipole hyperfine constant A , should then be rewritten to obtain the relative contact contribution to the hfa:

$${}^1\Delta_{exp}^2 = {}^1\Delta_c^2 \frac{A_c}{A} \quad (4)$$

where ${}^1\Delta_c^2$ is the hfa due to the contact interaction, that is for an s- or $p_{1/2}$ -electron. So far, we have considered direct interactions between the electron and the nucleus, but we should also include electron-electron interactions. One interaction, which can influence the hyperfine interaction, is the polarization of the electron core [10], which may give a substantial contribution to the experimental hfa [5]. Core polarization can be seen as an excitation of a d-electron, which will not itself give any contribution to the hfa, to an s-electron, which gives a large hfa. Since ${}^1\Delta_s^2$ is independent of n in the first approximation, it is possible to use it to obtain values of the core polarization [5,12].

From the discussion, one is led to the conclusion that one needs independent measurements of the nuclear magnetic moments and the A constants in order to obtain the hfa; however, this is not true. It has been shown by Persson [13] that it is possible to extract the anomaly solely from the A constants of two different atomic levels, provided the ratio $\left(\frac{A_s}{A}\right)$ differs substantially for the different levels. Comparing the A constant ratios, for two isotopes, at two atomic levels, gives:

$$\frac{A_B^{(1)} / A_B^{(2)}}{A_C^{(1)} / A_C^{(2)}} \approx 1 + {}^1\Delta_s^2 \left(\frac{A_s^B}{A^B} - \frac{A_s^C}{A^C} \right) \quad (5)$$

where B and C denote different atomic levels and 1 and 2 denote different isotopes. The ratio between the two A constant ratios for the isotopes will only depend on the difference of the contact contributions of the two atomic levels and the hfa for the s electron. It should be noted that the ratio $\left(\frac{A_s}{A}\right)$ is isotope independent. Once determined for one isotopic pair, the ratio can be used for all pairs, which is useful in the study of hfa in radioactive isotopes. The ratio can be determined in two different ways; either by making an analysis of the hyperfine interaction or by using a known hfa as a calibration. It should be pointed out that the atomic states used must differ significantly in the ratio $\left(\frac{A_s}{A}\right)$, as a small difference will lead to an increased sensitivity to errors, as can be deduced from Equation (5) [13].

Since the hfa is normally very small (1% or less), it is necessary to have high accuracy, better than 10^{-4} [5]. In the case of stable isotopes, there is no major problem to measure the nuclear magnetic dipole moment, with nuclear magnetic resonance (NMR) or atomic beam magnetic resonance (ABMR), while unstable isotopes are more difficult. In most cases, there does not exist any high precision

measurements of the nuclear magnetic dipole moment. However, there might exist measurements of two A constants, if the unstable isotopes' nuclear charge radius has been measured by means of laser spectroscopy [1]. In order to obtain the hfa, one needs to measure the A constants with an accuracy better than 10^{-4} . This can be done by laser spectroscopy when the A constant is larger than about 1000 MHz.

As pointed out in a review on hfa [5], off-diagonal hyperfine interactions may simulate an hfa. However, corrections due to off-diagonal hyperfine interaction affect mainly the electric quadrupole hyperfine interaction constants, unless the correction is very large, in which case the experimental error in the A constants is large. In most cases, the correction is smaller than the experimental error and can be neglected, especially with laser spectroscopy where the error is of the order of 1 MHz. In the present study, the experimental errors in the A constants are so large that off-diagonal hyperfine interaction corrections can be neglected.

Hyperfine Structure Measurements in Eu

Europium has been subject to many investigations since the measurements of Schüler and Schmidt in 1935 [14]. Since then, many measurements have been performed using a variety of different methods, for example; Fabry–Perot spectroscopy [15], atomic beam magnetic resonance [16], level crossing spectroscopy [17], laser atomic beam spectroscopy [18,19], laser-rfdouble resonance [20], and in an ion trap [21]. In total, the hfs has been determined in over 30 atomic states and 15 states in Eu^+ . The high accuracy in some measurements has made it possible to determine the hyperfine anomaly. One problem with a complex atom like Eu is that the hyperfine anomaly is state dependent and has to be analyzed to give the correct value [5]. A case study of the hyperfine anomaly was done by Büttgenbach in his review article [5], where he found the values of the s-electron hyperfine anomaly ($^{151}\Delta_s^{153}$) to be $-0.64(3)\%$, $-0.66(3)\%$, and $-0.59(5)\%$, depending on the states and experimental method used. The average value $-0.64(4)\%$ is probably a very good approximation, since the value obtained from Fabry–Perot measurements in the ion ($-0.59(5)\%$) is probably too low. The accurate measurements using an ion trap by Becker et al. [21] obtained an uncorrected, i.e., state dependent, value of $-0.663(18)\%$ for the hyperfine anomaly in the ground state of Eu^+ . The uncertainty in this value originates mainly from the nuclear magnetic dipole moment measurements [16]. The hfs in this state is not entirely due to contact interaction, but has a small non-contact interaction, as can be deduced from the experimental g_J factor. Since these contributions have the opposite sign, the state dependent hfa is slightly larger than the contact hfa.

There is no doubt that the high accuracy of the ion trap will give very precise values for the hyperfine anomaly, once the accuracy of the nuclear magnetic dipole moment can match the accuracy of the A constant ($\approx 10^{-8}$) in ion trap measurements [22] or if the A constant can be measured at another level, atomic or double-ionized, with matching accuracy.

In a paper by Asaga et al. [23], a theoretical study of the hyperfine anomaly in odd isotopes of Eu was performed. They addressed the question of the universality of the empirical Moskowitz–Lombardi formula [24]. In this article, the method of Persson [13] is applied to the measurements of Ahmad et al. [25] and Hühnermann et al. [26] in order to give an estimate on the validity of the Moskowitz–Lombardi formula in Eu. In addition, a calculation of the hyperfine anomaly using the particle-rotor model is presented.

2. Hyperfine Anomaly in Unstable Eu Isotopes

The unstable Eu isotopes have been studied using laser spectroscopy by Ahmad et al. [25], Hühnermann et al. [26], and by Dörschel et al. [27]. Measurements have also been performed by Enders et al. [28–30] using a Paul-trap, to obtain the hyperfine structure constants in the ground and first metastable states of Eu^+ . The high precision values from the ion trap measurements cannot, for the time being, be used to extract the hyperfine anomaly, as the s-electron contribution is almost the same at these levels [13].

2.1. Measurements Done by Hühnermann et al.

Hühnermann et al. [26] obtained information of the hyperfine anomaly by defining an angular factor $f(1, 2)$, where 1 and 2 are two different isotopes, which was found from a fit of the experimental A constants. The factor $f(1, 2)$ was defined as:

$${}^1\Delta_{\text{exp}}^2 = -c_s \cdot f(1, 2) / a^{(2)} \quad (6)$$

where $c_s = \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}$ and $a^{(2)}$ the experimental A constant for a state in Isotope 2. In this way, an experimental value of $f(151, 153) = -0.551(3)$ MHz was obtained [26].

The factor $f(1, 2)$ can be expressed using the normal nomenclature, with the experimental, i.e., state dependent, hyperfine anomaly ${}^1\Delta_{\text{exp}}^2$, as [13],

$${}^1\Delta_{\text{exp}}^2 = \frac{a_s}{a_{\text{exp}}} \cdot {}^1\Delta_s^2 \quad (7)$$

and the factor c_s as the total angular part of the contact interaction in the parametrization of the hfs [2], provided that the states investigated are purely LS-coupled. This leads to:

$$f(1, 2) = a_s^{(2)} \cdot {}^1\Delta_s^2. \quad (8)$$

Note that the (contact) factor $a_s^{(2)}$ must be corrected, so that it only takes into account the angular parts for the electrons influenced by the hyperfine anomaly, that is s-electrons. The states Hühnermann et al. studied, the $4f^7 5d^9 D_{J=2...6}$ in Eu^+ , are close to pure LS-coupling. However, there are no free s-electrons in this configuration, which is why the hyperfine anomaly arises from core polarization. The core polarization can be expressed as an excitation of an d-electron to an s-electron, thus showing a hyperfine anomaly. Since the hyperfine anomaly, due to core polarization, shows the same angular dependence as the contact interaction, one can use the hyperfine anomaly to find the core polarization [5,12]. The factor $a_s^{(2)}$ should then be the core polarization contribution to the hfs. An analysis of this case has been performed by Persson [12]. Using his value ($a_s^{(151)} = -669(40)$ MHz) and the angular part for the d-electron ($\frac{1}{8}$ of the total angular contribution), one finds that:

$$f(1, 151) = -669/8 \cdot {}^1\Delta_s^2. \quad (9)$$

yielding ${}^{151}\Delta_s^{153} = -0.659(4)(40)\%$, where the error is experimental and from the analysis, respectively, in agreement with other results. The values of $f(1, 2)$ for other isotopes from Hühnermann et al. [26] are given in Table 1 together with the derived hyperfine anomaly, ${}^1\Delta_s^2$. Note that the sign of $f(1, 2)$ was different in the original article, since the factor c_s was defined in a different way in this article.

Table 1. Hyperfine anomalies for Eu isotopes, obtained from the work of Hühnerman et al. [26]. Only experimental errors are given.

A	$f(151, A)$	${}^{151}\Delta_s^A$ (%)
145	0.07(20)	-0.08(24)
146	-0.11(37)	0.13(44)
147	-0.31(40)	0.37(48)
151	0	0.00
152	-0.42(5)	0.50(6)
153	0.551(3)	-0.659(4)

2.2. Measurements Done by Ahmad et al.

The values of Ahmad et al. [25] are used directly together with Equation (3) in order to obtain the hyperfine anomaly. In their study of nuclear spins, moments, and changes of the mean square

charge radii, they used two atomic transitions, 459.4 nm and 462.7 nm, connecting the atomic ground state ($4f^7 6s^2 \ ^8S_{7/2}$) with two excited states ($4f^7 6s 6p \ ^8P_{9/2,7/2}$). Since these transitions were studied with higher accuracy by Zaal et al. [18], there exists a calibration of the hyperfine anomaly for the stable isotopes.

The hfs of the ground state was not resolved in the measurements [25], which is why the A constants of the excited states were used. Using the measured A constants for the $^8P_{9/2,7/2}$ states from [25] and Equation (3), it is possible to deduce the hyperfine anomaly, for most unstable isotopes, using the $-0.64(4)\%$ value for the hyperfine anomaly between the stable isotopes as a calibration. The result is presented in Table 2. Comparing the results in Tables 1 and 2 shows an agreement within errors. The values for ^{147}Eu , still within errors, differ in sign, something that can be explained by the large experimental errors.

Table 2. Magnetic moments and hyperfine anomalies for Eu isotopes, obtained from Ahmad et al. [25].

A	I	μ_I	$^{151}\Delta_s^A (\%)$
142	1	1.536(19)	-0.14(1.18)
142m	8	2.978(11)	-0.08(31)
143	5/2	3.673(8)	-0.06(17)
144	1	1.893(13)	-0.19(48)
145	5/2	3.993(7)	-0.08(15)
146	4	1.425(11)	0.12(50)
147	5/2	3.724(8)	-0.12(17)
148	5	2.340(10)	0.08(31)
149	5/2	3.565(6)	-0.19(16)
150	5	2.708(11)	0.08(28)
151	5/2	3.4717(6)	0.00
153	5/2	1.5330(8)	-0.64(4)

As can be seen, the errors are larger than the actual values except for ^{149}Eu (Table 2) and ^{152}Eu (Table 1), which makes it difficult to draw any deeper conclusions, other than the general trends.

From the experimental values in Table 2, there seems to be an odd-even staggering, which changes sign at the $N = 82$ ($A = 145$) neutron shell closure, but the uncertainties are too large to be able to be sure about this. The drastic change in nuclear magnetic dipole moment between $N = 88$ ($A = 151$) and $N = 90$ ($A = 153$) due to the shape transition is also reflected in the hyperfine anomaly. The hfa for the lighter isotopes is fairly constant, indicating that the magnetization does not change much from the spherical ^{145}Eu nucleus to ^{151}Eu . With these experimental results, we can make comparisons with theoretical calculations.

3. Calculations of the Hyperfine Anomaly

The Bohr–Weisskopf effect, and thereby the hyperfine anomaly, was investigated by making particle-rotor calculations based on the modified oscillator (Nilsson) potential, using standard parameters [31] as much as possible. As the nuclear magnetic moment and the Bohr–Weisskopf effect calculations are mainly analogous, it is sensible to adjust the parameters in the calculation so that both the energy levels and nuclear magnetic moments fit well with the experimental values. The calculated hyperfine anomalies in the odd isotopes are given in Table 3. As expected, the Bohr–Weisskopf effect, and thus the hyperfine anomaly, stayed fairly constant from $A = 145$ to $A = 151$ with an abrupt change at the shape transition between $A = 151$ and 153.

For the odd isotopes, Asaga et al. [23] did a theoretical study and calculated the BW effect, and thus the hyperfine anomaly. Since they addressed the question on the validity of the empirical Moskowitz–Lombardi formula [24], which was used to estimate the hyperfine anomaly:

$$\epsilon_{BW} = \frac{\alpha}{\mu_I}, \tag{10}$$

the hyperfine anomaly was also calculated using this formula. The constant α was taken to be 0.015 n.m. It should be noted that this value of α is close to the value of Hg [24] and close to the values obtained for Ir [32,33] and Au [34]; however, the sign is different.

The results are presented in Table 3 together with the experimental values obtained from the measurements of Ahmad et al. [25].

Table 3. Hyperfine anomaly in Eu, experimental and calculations. MO denotes values obtained from Particle-rotor calculations, I and II values from [23], and ML values obtained from the Moskowitz–Lombardi formula [24].

	$^{151}\Delta_{\text{exp}}^A$ (%)	$^{151}\Delta_{MO}^A$ (%)	$^{151}\Delta_I^A$ (%)	$^{151}\Delta_{II}^A$ (%)	$^{151}\Delta_{ML}^A$ (%)
	Exp.	Calc.	Calc.	Calc.	Calc.
^{145}Eu	−0.08(15)	0.000	0.021	0.031	0.056
^{147}Eu	−0.12(17)	0.002	0.010	0.008	0.029
^{149}Eu	−0.19(16)	0.002	0.007	0.006	0.011
^{151}Eu	0	0.0	0	0	0.000
^{153}Eu	−0.64(4)	−0.768	−0.127	0.003	−0.546
^{155}Eu		−0.768	−0.127	−0.001	−0.555

Clearly, the theoretical calculations managed to reproduce the trend of the hyperfine anomaly, even if the values from Asaga et al. [23] were too small. It is interesting to note that the empirical Moskowitz–Lombardi (ML) formula seemed to be able to reproduce the hfa for Eu and not only for elements around $Z = 80$ (Ir,Au,Hg). However, the change in sign of α was an indication that this might be a coincidence.

If we inspect the actual values of the Bohr–Weisskopf effect (Table 4) instead of just looking at the hyperfine anomaly, we find a significant difference in the values obtained from the two methods of calculation. As we had an experimental value of the BW effect in ^{151}Eu from muonic X-ray [35] measurements, $\epsilon_{BW} = -0.63(13)\%$, it is possible to further discuss the methods. It is clear that the particle-rotor calculations showed a better agreement with experiment at least for the hyperfine anomaly. The BW effect was more difficult to calculate as there seemed to be an “offset” in the calculated results. The agreement with the experiment in Eu might be a coincidence, as preliminary calculations in Gd and Sm showed a more complex situation.

Table 4. Magnetic moments and Bohr–Weisskopf effect for odd Eu isotopes. MO denotes values obtained from particle-rotor calculations and I and II values from [23].

A	μ_I (exp)	μ_I (MO)	$\epsilon(\%)$ (MO)	$\epsilon(\%)$ I	$\epsilon(\%)$ II
145	3.993	3.773	−1.001	−1.067	−1.067
147	3.724	3.720	−1.003	−1.056	−1.044
149	3.565	3.552	−1.003	−1.053	−1.042
151	3.4717	3.506	−1.004	−1.046	−1.036
153	1.5330	1.532	−0.236	−0.919	−1.039
155	1.52	1.529	−0.236	−0.919	−1.035

The Empirical Moskowitz–Lombardi Formula

The empirical ML formula was established in 1973 as a rule for the s-electron BW effect in mercury isotopes [24].

$$\epsilon_{BW} = \frac{\alpha}{\mu_I}, \alpha = \pm 1.13 \cdot 10^{-2} \mu_N, I = l \pm \frac{1}{2} \tag{11}$$

where l is the orbital momentum for the odd neutron. It turned out that the empirical rule provided a better agreement with experimental hfa than the theoretical calculations performed by Fujita and Arima [4] using microscopic theory. The rule can be qualitatively explained by the microscopic theory used by Fujita and Arima [4], where the parameter α is more state independent than given by the theory. Further investigations gave an analogous expression for the odd-proton nuclei $^{191,193}\text{Ir}$, $^{197,199}\text{Au}$, and $^{203,205}\text{Tl}$, but also for the doubly-odd $^{196,198}\text{Au}$ nuclei. The results indicated that the spin operators $g_s^{(i)} \Sigma_i^{(1)}$ were state independent for these nuclei. It is worth noting that all nuclei discussed lied close to the doubly closed shell nucleus ^{208}Pb , where one would expect the single particle model to provide a good description of the nucleus. It is not apparent that the rule was applicable to lighter nuclei. Using the ML formula, the BW effect in ^{151}Eu attained a value of 0.325% compared to the experimental value of $-0.63(13)\%$ [35].

With the data presented here, it is possible to make a comparison with other lanthanide nuclei. As was shown in Eu, the ML formula seemed to account for the hfa, even though the obtained value of the BW effect differed more from the experimental value of ^{151}Eu . It should be noted that the sign of α was different from the value obtained for nuclei close to ^{208}Pb , indicating that the ML rule was not universal. In order to further test the ML formula, the values of α were deduced from the experimental values of the hfa and nuclear magnetic moments in Nd, Gd [36,37], and Eu, using:

$${}^1\Delta^2 = \alpha \left(\frac{1}{\mu_{I,1}} - \frac{1}{\mu_{I,2}} \right) \tag{12}$$

If the ML rule showed some sort of general validity, the values of α should stay fairly constant and show a different sign between Eu (odd-proton) and Nd and Gd (odd-neutron). The obtained values are shown in Table 5. As can be seen, there were no indications that the ML rule was applicable for these nuclei. The conclusion would be that one cannot use the ML rule for lighter nuclei. The experimental evidence indicated that the ML formula was only valid around $Z = 80$. The ML rule could not therefore be used when obtaining values for the nuclear magnetic dipole moment for unstable isotopes using hyperfine structure measurements.

Table 5. Hyperfine anomaly in the lanthanides.

	$\mu_{I,1}$	$\mu_{I,2}$	${}^1\Delta_s^2$ (%)	α (10^{-2})
$^{143,145}_{60}\text{Nd}$	-1.065	-0.656	0.2034	0.35
$^{151,153}_{63}\text{Eu}$	3.4717	1.533	-0.64	1.76
$^{157,155}_{64}\text{Gd}$	-0.3387	-0.2572	0.106	0.11

4. Conclusions

The method of Persson [13] was applied to Eu and provided preliminary values of the hyperfine anomaly, the experimental data were not precise enough for all isotopes. The values obtained were in agreement with the particle-rotor calculations and the theoretical predictions of the trends by Asaga et al. [23]. A comparison with the empirical ML formula indicated that it could be used for Eu; however, the constant α attained a different sign compared with the values for Ir, Au, and Hg, and a comparison with Nd and Gd showed that the ML formula is not applicable. This was clear evidence that the ML formula was not universal. There also seemed to exist an odd-even staggering of the hyperfine anomaly in Eu, similar to what was found in Au [34]. This analysis showed that there is a need for further studies of the hyperfine anomaly in Eu. The application of ion traps in measuring the A constants of unstable Eu isotopes [29,30] showed an excellent accuracy and will, when high accuracy measurements of the nuclear magnetic dipole moments are available or A constants in suitable atomic or ionic levels, give a deeper understanding of the hyperfine anomaly in Eu and hopefully to all nuclei.

As was shown in the case of Eu, it was possible to obtain information on the hfa without knowing the nuclear magnetic moment of the isotopes under study. It was also shown that the ML rule was not universal.

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