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Re-Evaluation of the Nuclear Magnetic Octupole Moment of ^{209}Bi

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Abstract: We modified the HFS92 code of the GRASP package in order to describe the magnetic octupole hyperfine interaction. To illustrate the utility of the modified code, we carried out state-of-the-art calculations of the electronic factors of the magnetic octupole hyperfine interaction constants for levels in the ground configuration of the Bi atom. The nuclear magnetic octupole moment of the ^{209}Bi isotope was extracted by combining old measurements of the hyperfine structures of $6p^3\ ^4S^o_{3/2}$ [Hull, R.; Brink, G. *Phys. Rev. A* **1970**, *1*, 685] and $^2P^o_{3/2}$ [Landman, D.A.; Lurio, A. *Phys. Rev. A* **1970**, *1*, 1330] using the atomic-beam magnetic-resonance technique with our theoretical electronic factors. The present extracted octupole moment was consistent with all the available values but the one obtained in the single-particle nuclear shell model approximation. This observation supports the previous finding that nuclear many-body effects, such as the core polarization, significantly contribute to the nuclear magnetic octupole moment in the case of ^{209}Bi.

Keywords: magnetic octupole hyperfine interaction; nuclear octupole moment; Bi; MCDHF method; GRASP package

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1. Introduction

Magnetic dipole moments are nowadays routinely measured to study nuclear structure, but magnetic octupole moments are comparatively studied a lot less. In the single-particle (SP) shell model, the nuclear magnetic octupole moment (Ω) can be obtained through [1]

$$\Omega_{\rm SP} = \mu_{\rm N}\frac{3}{2}\frac{(2I-1)}{(2I+4)(2I+2)}\langle r^2\rangle\begin{cases}(I+2)[(I-\frac{3}{2})g_l+g_s], & I=l+\frac{1}{2};\\ (I-1)[(I+\frac{5}{2})g_l-g_s], & I=l-\frac{1}{2},\end{cases} \quad (1)$$

where $\mu_{\rm N}$ is the nuclear magneton, I the nuclear spin, $\langle r^2\rangle$ the mean square radius of the radial distribution of the valence proton, and g_l and g_s are the orbital and spin gyromagnetic ratios, respectively. It was, however, shown that many-body effects among nucleons, such as the core polarization, modify this value considerably [2–5]. Moreover, as discussed by Sen'kov and Dmitriev [4], in the case of Bi, the octupole moment was more sensitive to these effects than the dipole moment. The same authors also pointed out that a systematic study of octupole moments would help to put constraints on the poorly known isoscalar part of the nuclear spin–spin interaction, as quoted in [6]. To date, a feasible and direct method to probe nuclear magnetic octupole moments, especially for radioactive nuclei, is to extract them from high-precision measurement of hyperfine structures. However, this

approach relies on accurate atomic structure calculations [5–9]. To this end, we modified the HFS92 code [10] to allow for the computation of the magnetic octupole hyperfine interaction and to deal with the Bohr–Weisskopf effect [11], using atomic electronic wave functions calculated with the GRASP2018 package [12]. Applying this new code, we made successful attempts to resolve the nuclear magnetic octupole moment puzzle in ^{173}Yb [13,14]. In the present paper, we show another application targeting the re-evaluation of the ^{209}Bi nuclear magnetic octupole moment.

2. Theory

The Hamiltonian of the magnetic octupole hyperfine interaction is written as [1,8]

$$\mathcal{H} = \mathbf{M}^{(3)} \cdot \mathbf{T}^{(3)}\,, \tag{2}$$

where $\mathbf{M}^{(3)}$ and $\mathbf{T}^{(3)}$ are spherical tensor operators of rank 3 in the electronic and nuclear spaces, respectively. This Hamiltonian leads in perturbation theory to the following first-order energy contribution to the hyperfine structure

$$\begin{aligned} E^{(1)} = \;& \langle \Upsilon\Gamma IJFM_F|\mathbf{M}^{(3)} \cdot \mathbf{T}^{(3)}|\Upsilon\Gamma IJFM_F\rangle \\ =\;& (-1)^{F-I-J}\sqrt{(2I+1)(2J+1)}\begin{Bmatrix} I & I & 3 \\ J & J & F \end{Bmatrix}\langle \Upsilon I||\mathbf{M}^{(3)}||\Upsilon I\rangle\langle \Gamma J||\mathbf{T}^{(3)}||\Gamma J\rangle\,. \end{aligned} \tag{3}$$

Here, the convention by Brink and Satchler [15] is adopted for the reduced matrix elements. $|\Upsilon\Gamma IJFM_F\rangle$ is the coupled wave function of the nuclear $|\Upsilon IM_I\rangle$ and electronic $|\Gamma JM_J\rangle$ components, in which the total angular momentum $\boldsymbol{F}$ is the vector sum of nuclear ($\boldsymbol{I}$) and electronic ($\boldsymbol{J}$) angular momenta. M_I, M_J, and M_F stand for the z-component of the corresponding angular momenta, and Υ and Γ are the other necessary quantum numbers characterizing the nuclear and electronic states, respectively.

Introducing the coefficient Ξ,

$$\Xi(I,J,F,k) = \frac{(-1)^{F-I-J}\begin{Bmatrix} I & I & k \\ J & J & F \end{Bmatrix}}{\begin{pmatrix} I & k & I \\ -I & 0 & I \end{pmatrix}\begin{pmatrix} J & k & J \\ -J & 0 & J \end{pmatrix}}\,, \tag{4}$$

Equation (3) factorizes as

$$\langle \Upsilon\Gamma IJFM_F|\mathbf{M}^{(3)} \cdot \mathbf{T}^{(3)}|\Upsilon\Gamma IJFM_F\rangle = \Xi(I,J,F,3)\langle \Upsilon II|M_0^{(3)}|\Upsilon II\rangle\langle \Gamma JJ|T_0^{(3)}|\Gamma JJ\rangle\,. \tag{5}$$

The magnetic octupole hyperfine interaction constant, C, is defined as

$$\begin{aligned} C \equiv\;& \langle \Upsilon II|M_0^3|\Upsilon II\rangle\langle \Gamma JJ|T_0^3|\Gamma JJ\rangle \\ =\;& -\Omega\langle \Gamma JJ|T_0^3|\Gamma JJ\rangle\,, \end{aligned} \tag{6}$$

where Ω is related to the nuclear magnetic octupole moment

$$\Omega = -\langle \Upsilon II|M_0^{(3)}|\Upsilon II\rangle\,. \tag{7}$$

The constant C can be deduced from high-precision hyperfine structure measurements. The value of the octupole moment Ω can be extracted from C if the electronic factor, $-C/\Omega$, that is, the magnetic field gradient at the nucleus produced by electrons in the atom [16,17],

$$\begin{aligned} -C/\Omega =\;& \langle \Gamma JJ|T_0^3|\Gamma JJ\rangle \\ =\;& \sqrt{\tfrac{J(J-1)(2J-1)}{(J+1)(J+2)(2J+3)}}\langle \Gamma J||\mathbf{T}^{(3)}||\Gamma J\rangle \\ =\;& \sqrt{\tfrac{J(J-1)(2J-1)}{(J+1)(J+2)(2J+3)}}\langle \Gamma J||\textstyle\sum_{\nu=1}^{N}\frac{-\mathrm{i}\,\alpha}{3r_\nu^4}\boldsymbol{\alpha}_\nu \cdot (\boldsymbol{L}C^{(3)}(\theta_\nu,\phi_\nu))||\Gamma J\rangle \end{aligned} \tag{8}$$

is provided by atomic structure calculations. In Equation (8), α is the fine-structure constant, $\boldsymbol{\alpha}$ the Dirac matrix, r the radial coordinate, $\boldsymbol{L}C^{(k)}(\theta,\phi)$ the transverse vector spherical harmonic [18], and the summation runs over the N electrons. The accuracy of such atomic structure calculations relies on the quality of the electronic wave functions. In this work, the latter were expanded in jj-coupled configuration state functions (CSFs) determined using the GRASP2018 package [12] based on the multiconfiguration Dirac–Hartree–Fock (MCDHF) method [19,20].

3. Results

The electronic factors, $-C/\Omega$, entering in the magnetic octupole hyperfine interaction constant, C, were estimated for the $^4S^o_{3/2}$, $^2D^o_{3/2,5/2}$ and $^2P^o_{3/2}$ levels of the bismuth [Kr]$4d^{10}4f^{14}5s^25p^65d^{10}6s^26p^3$ ground configuration. The results for various computational models are presented in Table 1. For comparison, the electronic factors, $A_{\rm el} = A\ (I/\mu)$, of the magnetic dipole hyperfine interaction constants A are also reported in this table. The details on our correlation models can be found in [21,22]. The computational strategy had two components. The first one was to optimize a set of one-electron orbitals based on an approach combining the MCDHF method and the second-order perturbation theory. The second one was to construct a configuration space expanded by CSFs and perform relativistic configuration interaction (RCI) computations to capture electron correlation effects. The CSFs were generated by substituting one, two, three, etc., and/or even all the electrons from the occupied orbitals, in the reference configuration to selected orbitals in the orbital basis. Note that no approximation such as the second-order perturbation was made in our RCI computations. The scope of the electron correlation effects which could be considered depended to a large extent on the available computational resources. For the present case of Bi, we accounted for the electron correlation within the $\{6s, 6p\}$ valence shells (valence correlation) and between the $n \geq 4$ core-shell and the valence-shell electrons (core-valence correlation). The CSFs generated by selecting triple and quadruple substitutions from the valence shells were also included into the configuration space. This model was referred to as "MR-CI". The $n = 3$ core-valence correlation effects (CV_3) were also investigated on the basis of MR-CI calculations. The final results, labeled as "+Breit", indicated that the Breit interaction was added to the Dirac–Coulomb Hamiltonian within the CV_3 model.

Table 1. Electronic factors of the magnetic dipole ($A_{\rm el}$ in MHz/$\mu_{\rm N}$) and magnetic octupole ($-C/\Omega$ in kHz/($\mu_{\rm N}\times$b)) hyperfine interaction constants of states in the [Kr]$4d^{10}4f^{14}5s^25p^65d^{10}6s^26p^3$ ground configuration of Bi.

Models	$^4S^o_{3/2}$		$^2D^o_{3/2}$		$^2D^o_{5/2}$		$^2P^o_{3/2}$	
	$A_{\rm el}$	$-C/\Omega$	$A_{\rm el}$	$-C/\Omega$	$A_{\rm el}$	$-C/\Omega$	$A_{\rm el}$	$-C/\Omega$
DHF	41.9	−22.5	−530	14.6	2954	68.9	883	−33.4
MR-CI	−572	−32.4	−1357	24.1	2691	83.2	13,245	−41.7
CV_3	−540	−32.9	−1370	24.5	3017	84.4	13,427	−42.3
+Breit	−554	−32.6	−1338	24.2	3007	84.3	13,365	−42.2

Comparing the CV_3 results with those obtained in the Dirac–Hartree–Fock (DHF) approximation, we see from Table 1 that the electron correlation effects affected both the magnetic dipole and octupole hyperfine interaction constants. It should be stressed that the $n = 3$ core-valence correlation effect on the magnetic dipole hyperfine interaction constants was not negligible, although the $3s$, $3p$, and $3d$ orbitals were located close to the nucleus. However, the magnetic octupole hyperfine interaction constants were not as sensitive to the electron correlation effects as the dipole hyperfine interaction. This could possibly be attributed to the differences in the radial dependence of magnetic dipole and octupole interactions, or to different tensorial structures thereof. The magnetic octupole hyperfine interaction is proportional to $1/r^4$ (see Equation (8)), while the dipole interaction

is proportional to $1/r^2$ [10,23]. It means that the radial integrals over the region closer to the nucleus, where the Coulomb interaction between the nucleus and electrons dominates, yield relatively larger contribution to the magnetic octupole hyperfine interaction constant, compared to the magnetic dipole constant. As a result, we could utilize the computational uncertainty of $A_{\rm el}$ as an upper limit of the theoretical uncertainty on $-C/\Omega$. In the case of Bi, this was estimated to be about 10% by comparing our A constants with measurements.

The hyperfine structures of the ^{209}Bi ground configuration $6p^3$ were measured by Hull and Brink [24], and Landman and Lurio [25] for $^4S^o_{3/2}$ and $^2P^o_{3/2}$, respectively, using the atomic-beam magnetic-resonance technique. As discussed by these authors, it is necessary to examine in detail the fine and hyperfine structures of all the $6p^3$ levels to properly interpret the measurements. The "corrections" they introduced take into account the second-order energy contributions due to the hyperfine coupling with the other fine-structure levels that significantly affect the extracted experimental C constants from the measured hyperfine intervals. Combining these "corrected" C constants with our electronic factor $-C/\Omega$ values evaluated with the "+Breit" model, we extracted the nuclear magnetic octupole moments of ^{209}Bi. Taking the arithmetic average over the two octupole moment values so determined, i.e., $\Omega = 0.56\ \mu_{\rm N} \times$ b for $^4S^o_{3/2}$, and $\Omega = 0.46\ \mu_{\rm N} \times$ b for $^2P^o_{3/2}$, we obtained $\Omega(^{209}{\rm Bi}) = 0.51\ \mu_{\rm N} \times$ b.

These nuclear magnetic octupole moment values are listed in Table 2 and compared with the $\Omega_{\rm SP}$ value estimated from the SP shell model (see Equation (1)). The ^{209}Bi isotope has an unpaired $h_{9/2}$ proton and according to Equation (1),

$$\Omega_{\rm SP} = 0.417 \mu_{\rm N} \langle r^2 \rangle = 0.127 \mu_{\rm N} \times {\rm b}\,, \tag{9}$$

where we used $g_l = 1$, $g_s = 5.58$, and the root-mean-square value of the nuclear radius $\langle r^2 \rangle^{1/2} = 5.5211$ fm [26]. Besides our set of Ω values, we display in the same table the values extracted by Hull and Brink [24] and Landman and Lurio [25] combining their C experimental values with their original electronic factors $-C/\Omega$. As it can be observed, these values were consistent with ours, which was a bit surprising when considering the simplicity of Schwartz's Equation [1,27] that they used for relating their measured hyperfine constant with the nuclear magnetic octupole moments (see Equation (7) of [24] or (14a,b,c) of [25]).

More recently, considering the nuclear core polarization and the correction to the nuclear electromagnetic current due to velocity-dependent interactions, Sen'kov and Dmitriev reported the magnetic octupole moment $\Omega = 0.48\ \mu_{\rm N} \times$b from their nuclear structure calculation [4]. This value agreed reasonably well with other results, except for $\Omega_{\rm SP}$. This observation confirmed the significance of many-body interactions among nucleons to the octupole moment of ^{209}Bi. It is interesting to note that the SP shell model also performed poorly for the magnetic dipole moment of ^{209}Bi [4]. It seems that the magnetic properties of ^{209}Bi cannot be understood using a pure SP shell model.

Table 2. Nuclear octupole moments of ^{209}Bi in $\mu_{\rm N} \times$b units. $\Omega_{\rm SP}$ was obtained with Equation (1) using $\langle r^2 \rangle^{1/2} = 5.5211$ fm [26]. Numbers in parentheses reflect uncertainties. NS stands for the nuclear structure calculation.

$\Omega_{\rm SP}$	This Work			Others		
	$^4S^o_{3/2}$	$^2P^o_{3/2}$	Average	$^4S^o_{3/2}$ [24]	$^2P^o_{3/2}$ [25]	NS [4]
0.127	0.56	0.46	0.51(5)	0.43	0.55(3)	0.48

4. Conclusions

We carried out ab initio calculations of the electronic factors relevant to the magnetic octupole hyperfine interaction constants of the levels in the ground configuration of Bi by using an extended version of the HFS92 code in the framework of the MCDHF method. Based on these electronic factors, the nuclear magnetic octupole moment of the ^{209}Bi isotope

was extracted from the experimental C constants. Our octupole moment values agreed well with other estimations, including the most recent nuclear structure calculations [4]. The discrepancies with the Ω_{SP} value confirmed the significance of the many-body interactions in the description of this nuclear property. The present study should motivate nuclear physicists to investigate the magnetic octupole moments of radioactive isotopes along isotope chains and probe the nuclear many-body effects. The new development of the HFS92 code and the GRASP package provide useful and promising tools for this purpose.

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