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Laser-rf double-resonance spectroscopy of ^{177,179}Hf

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The atomic-beam laser-rf double-resonance technique has been used to make precision measurements of the hyperfine structure (hfs) splittings in the $5d^26s^2\,{}^3F_J$ ($J=2$–4) states in ^{177,179}Hf I. By making corrections to the second-order hfs effects, magnetic dipole (A), electric quadrupole (B), and magnetic octupole (C) hfs constants are obtained for the ^3F_J states in ^{177,179}Hf: The constant C has a strong J dependence as well as does A and B; the isotopic ratio of the constant A between ^{177}Hf and ^{179}Hf has a significant J dependence, indicating J-dependent hyperfine anomalies, while that of the constant C is independent of J within experimental uncertainties, which is taken to be the ratio of the nuclear-ground-state magnetic octupole moments Ω, i.e., $\Omega(^{177}\mathrm{Hf})/\Omega(^{179}\mathrm{Hf})=-0.41(9)$. It is also pointed out that there is a clear indication of nuclear-spin dependence of the isotopic ratios of C. The single-electron hfs parameters of the $5d$ electron for the $5d^26s^2$ configuration in Hf I are obtained, and the Sternheimer shielding factor $R_{5d}=-0.397(8)$ is derived.

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I. INTRODUCTION

The only two odd-mass stable hafnium isotopes are ^{177}Hf and ^{179}Hf with nuclear spins $I=\frac{7}{2}$ and $\frac{9}{2}$, respectively. The atomic ground state is $5d^26s^2\,{}^3F_2$, and there are low-lying states $^3F_3$ and $^3F_4$. As is commonly the case with the $5d$ elements, the hafnium atom has many low-lying levels below 3×10^4 cm^{-1} that are members of the odd-parity configurations as well as the even-parity ones [1]. Probable mixings from the open s shell of $5d^36s$ may lead to significant hyperfine anomalies as well as configuration-interaction effects in the ground-state configuration $5d^26s^2$. Detailed optical studies of the hyperfine structure (hfs) of ^{177}Hf and ^{179}Hf will provide key information to analyze hyperfine spectroscopic data on other Hf isotopes not only from the atomic spectroscopic point of view but also from the nuclear spectroscopic point of view. The Sternheimer shielding effect of the core electrons [2,3], for example, must be evaluated precisely. They will also be a good example to study the dependence of hfs constants on nuclear spin I as well as electronic angular momentum J.

For Hf, only a few hfs measurements have been reported so far; this is because Hf is a refractory element whose atomic beams are difficult to produce stably. Büttgenbach and co-workers [4,5] reported hfs measurements for the ground state $5d^26s^2\,{}^3F_2$ in ^{177,179}Hf and later [6] for the $5d^26s^2\,{}^3F_3$ and $^3F_4$ states in ^{179}Hf by means of the atomic-beam magnetic resonance (ABMR) technique. Hyperfine structures for the three optical transitions have recently been measured using the laser-ablation atomic beam [7,8] and the resonance gas cell methods [9]. Very recently, Zimmermann *et al.* reported hfs and isotope shift measurements of 14 transitions from the $5d^26s^2$ configuration by means of laser spectroscopy using an electron bombardment technique [10]. We have confirmed their data on hfs and isotope shift by using our powerful and stable atomic-beam source [11,12].

Hyperfine structures in Hf I are, however, not well understood even for the ground-state term ^3F, and no data are available for the $5d^26s^2\,{}^3F_3$ and $^3F_4$ states in ^{177}Hf. The nuclear quadrupole moments of ^{177,179}Hf obtained by Büttgenbach *et al.* [5] deviate largely from the values of muonic x-ray measurements [13], and the Sternheimer shielding factor of ^{177,179}Hf obtained by Tanaka *et al.* [14] seems unreasonably large compared to those of ^{191,193}Ir, another $5d$ element, and those of rare-earth ele-

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ments [14].

In this paper, we shall report high-precision hfs measurements for the $5d^26s^2$ configuration in ^{177,179}Hf I using the laser-rf double-resonance (LRDR) method pioneered by Ertmer and Hofer [15], which enable us to discuss the configuration mixing, the Sternheimer shielding effect, the hyperfine anomaly, and nuclear quadrupole and octupole moments in detail.

II. EXPERIMENTAL METHOD

The present experiment was carried out by means of the LRDR method. The Hf atomic beam was produced using the Ar-ion-sputtering method [11,12]. The Ar-ion beam generated by an electron gun was accelerated to 8 keV and focused to about 1 mm^2 on the Hf natural metallic target: the isotopic compositions are 0.16% ^{174}Hf, 5.2% ^{176}Hf, 18.6% ^{177}Hf, 27.1% ^{178}Hf, 13.8% ^{179}Hf, and 35.2% ^{180}Hf.

The laser beam from a cw ring dye laser (Coherent 699-29) that was optically pumped by an Ar-ion laser (Spectra Physics 171-19) with rhodamine 110 was split into two beams, i.e., pumping and probe lasers. To reduce the Doppler broadening, both the atomic beam and the probe laser were collimated and the pumping and probe lasers crossed the atomic beam perpendicularly. Fluorescences induced by the probe laser were collected and focused on a cooled single photon-counting photomultiplier (Hamamatsu R1333) with a spherical mirror.

A 3-cm-long wire (rf loop) was placed between the pumping and probe lasers parallel to the atomic beam to produce an rf field. The ratio frequency was generated by a synthesizer (HP8341B), and the rf power applied was less than 30 mW at the terminal connected to the wire, which was far below the level that gave rise to power broadenings and shifts. To cancel the stray magnetic field in the rf region, three pairs of Helmholtz coils were used in three dimensions. The experimental setup and the way of LRDR measurements work have been described in detail previously [16,17].

III. EXPERIMENTAL RESULTS

Figure 1 shows three transitions and related energy-level schemes studied in this experiment. The first transition is from the ground state $5d^26s^2\,{}^3F_2$; and the second $^3F_3$ at 2356.68 cm^{-1} and third $^3F_4$ at 4567.64 cm^{-1} are both transitions associated with the excited states of the $5d^26s6p$ configuration [1]. Hyperfine structure components of the 555.06-, 537.39-, and 545.29-nm optical transitions were used for the LRDR method to precisely determine the hfs splittings in $^3F_2$, $^3F_3$, and $^3F_4$, respectively.

First, we carried out laser-induced fluorescence measurements for the three transitions by scanning the laser frequency without the pumping laser. A typical spectrum measured for the 555.06-nm transition is shown in Fig. 2. The observed linewidth of a single hfs peak was about 35 MHz at full width at half maximum (FWHM).

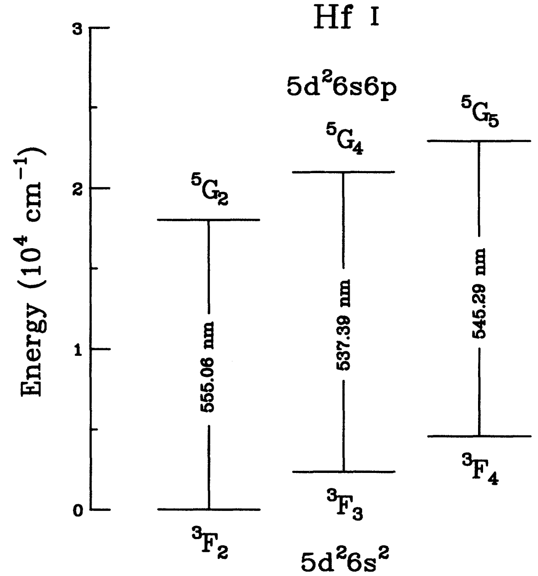

FIG. 1. Transitions studied and related energy-level schemes in Hf I.

On the basis of the present optical measurements, we carried out the measurement of LRDR for the ^3F_J states in ^{177,179}Hf I. The hfs splittings of the lower and upper states of the 555.06-nm transition are shown in Figs.3(a) and 4(a) for ^{177}Hf and ^{179}Hf, respectively. The lower $^3F_2$ states have four hfs splittings to be determined for both ^{177}Hf and ^{179}Hf. A typical rf-resonance spectrum observed for ^{177}Hf is shown in Fig. 3(b) for the hfs splitting $F-F'=\frac{5}{2}-\frac{7}{2}$ in $^3F_2$; the laser frequency was fixed at the hfs transition $\frac{5}{2}$ ($^3F_2$)$-\frac{7}{2}$ ($^5G_2$), as shown in Fig. 3(a), where F is the total angular momentum of the atom. A typical rf-resonance spectrum observed for ^{179}Hf is shown

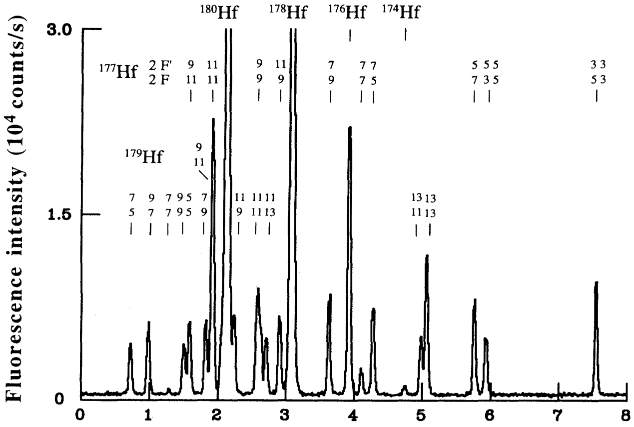

FIG. 2. Laser-induced fluorescence spectrum of the 555.06-nm ($^3F_2$–$^5G_2$) transition in Hf I. Peaks of even-mass isotopes are labeled with isotopic symbols. The hfs peaks of odd-mass isotopes ^{177}Hf and ^{179}Hf are labeled with a pair of the total angular momenta F (the lower state) and F' (the upper site) [see Figs. 3(a) and 4(a)].

in Fig. 4(b) for the hfs splitting $F-F'=\frac{11}{2}-\frac{13}{2}$ in $^3F_2$; the laser frequency was fixed at the hfs transition $\frac{11}{2}$ ($^3F_2$)$-\frac{13}{2}$ ($^5G_2$), as shown in Fig. 4(a). The observed resonance peak width (FWHM) is about 140 kHz for ^{177}Hf [Fig. 3(b)] while it is about 120 kHz for ^{179}Hf [Fig. 4(b)]; these resonance widths are mainly from the limited transit time of atoms in the rf field and also from the residual Zeeman broadening caused by the remnant magnetic field in the rf region [16]. The discrepancy between the peak widths for ^{177}HF and ^{179}Hf is due to the different residual Zeeman broadening caused by the different nuclear spin and angular momentum F of the related hfs levels.

By tuning the laser frequency to other hfs transitions [see Figs. 3(a) and 4(a)], the rf-resonance spectra were measured for all hfs splittings in ^3F_J ($J=2$–4) of ^{177,179}Hf, except the hfs splitting $F-F'=\frac{1}{2}-\frac{3}{2}$ in $^3F_4$ for ^{179}Hf whose corresponding fluorescence peak was too weak. The zero-field hfs splittings were thus determined with great precision by making a least-squares fit of a Lorentzian function to the experimental rf-resonance spectrum. Table I lists the determined hfs splittings. Previous values for the $^3F_2$ state measured by the ABMR [5] are also given for comparison. The sign of the hfs splittings (level ordering) was determined from the fluorescence spectra measured because the rf measurement is completely insensitive to the sign.

TABLE I. Zero-field hfs splittings $\Delta\nu$ measured by the LRDR method for the ^3F_J states in ^{177,179}Hf I. Previous values for the $^3F_2$ state are listed for comparison. Columns 4 and 6 give the difference between observed frequencies corrected for second-order hfs effects (Expt.) and calculated ones (Fit) using the corrected hfs constants in Table II for ^{177}Hf and ^{179}Hf, respectively.

		^{177}Hf		^{179}Hf	
State	$F-F$	$\Delta\nu$ (kHz)	(Expt.$-$Fit) (kHz)	$\Delta\nu$ (kHz)	(Expt.$-$Fit) (kHz)
$^3F_2$	$\frac{3}{2}-\frac{5}{2}$	4 862.89(31)	0.25		
		4 863.56(32)[a]			
	$\frac{5}{2}-\frac{7}{2}$	162 887.27(35)	-0.56	$-558\,670.75(61)$	0.68
		162 886.85(15)[a]		$-558\,671.74(24)$[a]	
	$\frac{7}{2}-\frac{9}{2}$	447 008.41(25)	0.14	$-541\,909.48(55)$	-1.04
		477 008.47(21)[a]		$-541\,910.44(7)$[a]	
	$\frac{9}{2}-\frac{11}{2}$	991 792.28(38)	-0.04	$-392\,848.44(36)$	0.32
		991 792.02(24)[a]		$-392\,847.75(37)$[a]	
	$\frac{11}{2}-\frac{13}{2}$			$-82\,131.15(25)$	-0.02
				$-82\,132.14(60)$[a]	
$^3F_3$	$\frac{1}{2}-\frac{3}{2}$	$-26\,074.62(95)$	-0.42		
	$\frac{3}{2}-\frac{5}{2}$	$-4\,224.51(85)$	-1.30	$-320\,992.3(20)$	-0.61
	$\frac{5}{2}-\frac{7}{2}$	76 482.36(66)	0.07	$-395\,075.3(21)$	1.05
	$\frac{7}{2}-\frac{9}{2}$	239 588.76(50)	1.08	$-414\,844.4(10)$	0.26
	$\frac{9}{2}-\frac{11}{2}$	508 632.03(48)	-0.94	$-364\,776.8(12)$	-1.07
	$\frac{11}{2}-\frac{13}{2}$	907 138.88(65)	0.40	$-229\,354.0(12)$	-0.75
	$\frac{13}{2}-\frac{15}{2}$			6 916.5(10)	-0.11
$^3F_4$	$\frac{1}{2}-\frac{3}{2}$	$-77\,362.7(11)$	1.17		
	$\frac{3}{2}-\frac{5}{2}$	$-92\,390.41(81)$	1.06	$-337\,533.8(22)$	-0.43
	$\frac{5}{2}-\frac{7}{2}$	$-52\,595.46(80)$	-1.30	$-421\,949.0(23)$	1.94
	$\frac{7}{2}-\frac{9}{2}$	63 960.32(70)	-0.67	$-445\,771.3(17)$	-0.80
	$\frac{9}{2}-\frac{11}{2}$	279 208.08(79)	1.46	$-424\,533.4(17)$	0.35
	$\frac{11}{2}-\frac{13}{2}$	615 069.12(80)	-0.47	$-313\,785.7(23)$	-1.54
	$\frac{13}{2}-\frac{15}{2}$	1 093 465.0(15)	0.15	$-109\,069.4(20)$	1.26
	$\frac{15}{2}-\frac{17}{2}$			204 047.0(18)	-0.29

[a] Reference [5].

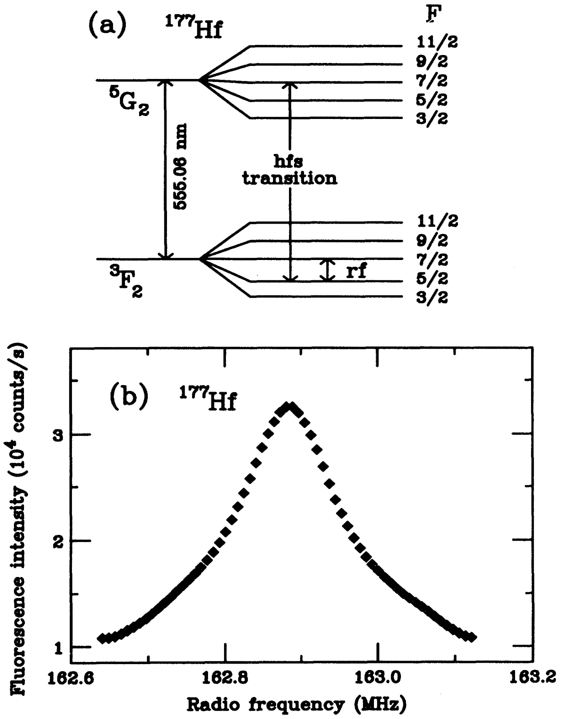

FIG. 3. hfs splitting schemes of ^{177}Hf (a) and measured rf-resonance spectrum for the zero-field hfs splitting $F-F'=\frac{5}{2}-\frac{7}{2}$ in $^{3}F_2$ of ^{177}Hf (b).

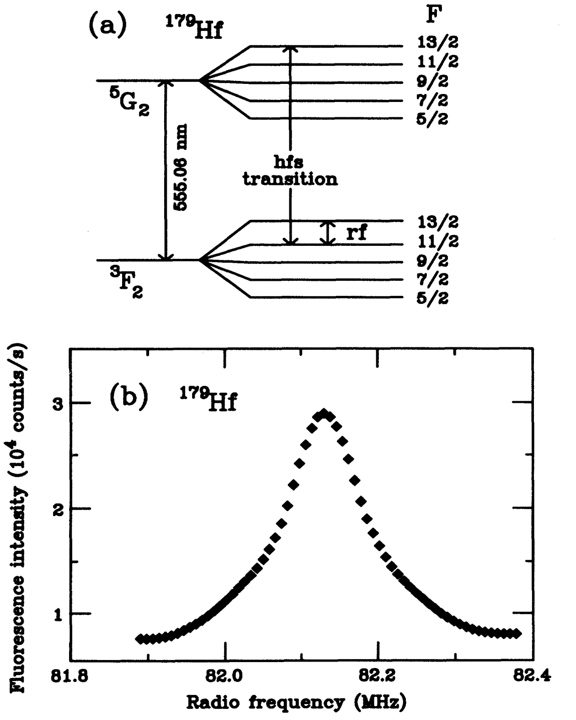

FIG. 4. hfs splitting schemes of ^{179}Hf (a) and measured rf-resonance spectrum for the zero-field hfs splitting $F-F'=\frac{11}{2}-\frac{13}{2}$ in $^{3}F_2$ of ^{179}Hf (b).

IV. ANALYSIS AND DISCUSSION

A. hfs constants *A*, *B*, and *C* for the $5d^2 6s^2\,{}^3F_J$ ($J=2-4$) states

Since the LRDR measurement yields the hfs splitting precise enough for correction of the order of kHz to be taken into account, the correction for second-order hfs effects should be made to determine hfs constants from the observed hfs splittings. The energy shift of a hfs state due to the second-order hfs effects from other hfs states with the same angular momentum *F* is given by

$$\delta E(\varphi,IF)=\sum_{\varphi'\neq\varphi}\frac{|\langle\varphi,IF|H_{\rm hfs}|\varphi' IF\rangle|^2}{E_\varphi-E_{\varphi'}}\,, \tag{1}$$

where $H_{\rm hfs}$ denotes the hfs Hamiltonian, $|\varphi,IF\rangle$ the wave function of the hfs state, and φ and E_φ the wave function and energy of the fine-structure state, respectively The matrix element $\langle\varphi,IF|H_{\rm hfs}|\chi' IF\rangle$ was given by Childs [18] for the pure *LS* coupling scheme of the l^N configuration.

To calculate the matrix element $\langle\varphi,IF|H_{\rm hfs}|\varphi',IF\rangle$, the single-electron hfs parameters $a_{nl}^{k_s k_l}$ for the magnetic interaction and $b_{nl}^{k_s k_l}$ for the electric interaction [18,19] have to be evaluated. For the $5d^2 6s^2$ configuration, the experimental magnetic-dipole hfs constants *A* and electric-quadrupole ones *B* are written as [20]

$$A(J)=\alpha_{5d}^{01}(J)a_{5d}^{01}+\alpha_{5d}^{12}(J)a_{5d}^{12}+\alpha_{5d}^{10}(J)a_{5d}^{10}\,, \tag{2}$$

$$B(J)=\beta_{5d}^{02}(J)b_{5d}^{02}+\beta_{5d}^{13}(J)b_{5d}^{13}+\beta_{5d}^{11}(J)b_{5d}^{11}\,, \tag{3}$$

where $\alpha_{5d}^{k_s k_l}$ and $\beta_{5d}^{k_s k_l}$ are the angular coefficients [21].

First, using the observed hfs splittings in Table I, the magnetic dipole (*A*), electric quadrupole (*B*), the magnetic octupole (*C*) hfs constants were evaluated approximately, the results for the $^3F_J(J=2-4)$ states in ^{177,179}Hf are given in Table II (uncorrected) in which the previously reported values [5,6] for $^3F_2$ in ^{177}HF and ^3F_J ($J=2-4$) in ^{179}Hf are included for comparison. The values of *A* and *B* were used to obtain the single-electron hfs parameters $d_{5d}^{k_s k_l}$ and $b_{5d}^{k_s k_l}$. The observed hfs splittings in ^3F_J (Table I) were thus corrected for second-order hfs perturbations, where the pure *LS* coupling scheme was used and perturbation by the other hfs state within the $5d^2 6s^2$ configuration was considered. The hfs constants *A*, *B*, and *C* were obtained from the corrected hfs splittings (Expt.) and the results are also listed in Table II (corrected) for the ^3F_J ($J=2-4$) states in ^{177,179}HF, and the previous corrected values [5,6] are included as well for comparison. From Table I, it is seen that the present measured hfs splittings of ^{177,179}Hf for $^3F_2$ are slightly different from the previous measurements [5] to such an extent that the uncorrected *A*, *B*, and *C* values deviate from the previous ones (see Table II). This is probably because we measured hfs splittings in the zero field; they extracted the splittings from the Zeeman splitting measurements (ABMR) [5]. It should also be pointed out that the correction for the second-order hfs effects δE should have been different from that in the previous

TABLE II. Magnetic dipole (A), electric quadrupole (B), and magnetic octupole (C) hfs constants determined by the LRDR method for the $5d^2 6s^2\,3F_J$ states in ^{177}Hf and ^{179}Hf. Uncorrected constants are those obtained from the observed hfs splittings; corrected constants are those obtained from the hfs splittings corrected for the second-order hfs effects.

State	hfs constants	Uncorrected			Corrected		
		$^3F_2$	$^3F_3$	$^3F_4$	$^3F_2$	$^3F_3$	$^3F_4$
^{177}Hf	A (MHz)	113.433 48(4)	80.706 77(6)	69.039 97(7)	113.433 29(4)	80.706 64(6)	69.039 73(7)
		113.433 31(3)[a]			113.433 14(7)[a]		
	B (MHz)	624.333 19(42)	823.954 6(11)	1432.783 3(20)	624.330 13(42)	823.949 4(11)	1432.773 5(20)
		624.334 37(30)[a]			624.329 3(13)[a]		
	C (kHz)	−0.38(3)	−1.28(9)	−0.78(19)	0.17(3)	0.23(9)	0.50(19)
		−0.37(3)[a]			0.27(18)[a]		
^{179}Hf	A (MHz)	−71.428 48(4)	−50.804 84(10)	−43.456 36(13)	−71.428 84(4)	−50.804 91(10)	−43.456 37(13)
		−71.428 67(4)[a]	−50.806 1(17)[b]	−43.456 3(15)[b]	−71.428 91(9)[a]	−50.806 1(17)[b]	−43.456 3(15)[b]
	B (MHz)	705.524 87(61)	931.103 5(24)	1619.110 3(42)	705.518 25(61)	931.090 9(24)	1619.097 1(42)
		705.526 92(56)[a]	931.113(42)[b]	1619.128(55)[b]	705.518 1(24)[a]	931.100(42)[b]	1619.100(55)[b]
	C (kHz)	−1.19(5)	−1.81(25)	−2.51(50)	−0.41(5)	−0.60(25)	−1.19(50)
		−1.16(5)[a]			−0.43(20)[a]		

[a] Reference [5].
[b] Reference [6].

study [5] because of the Zeeman splittings. This is considered to be the reason why the errors of the present corrected A, B, and C constants for $^3F_2$ are significantly smaller than those of the previous ones [5] (see Table II).

To check the second-order hfs corrections, the hfs splittings were recalculated using the corrected hfs constants A, B, and C listed in Table II and, the differences between the experimental hfs splittings corrected for the second-order hfs effects (Expt.) and calculated ones (fit) were obtained as listed in Table I for ^3F_J ($J=2$–4) in ^{177,179}Hf. It is seen from Table I that the differences are very small, being around 1 kHz, the same order of magnitude of uncertainties. It should be pointed out, however, that the differences uncorrected for second-order hfs perturbations are large, being 1–6 kHz. This shows that the corrections for the second-order hfs effects are important in precision measurements. It is also found from Table II that the corrections have small but significant effects of the order of $(2-10)\times10^{-6}$ on the hfs constants A and B while so remarkable effects (50–160 %) on the constant C.

Although Büttgenbach *et al.* [5] used four parameters A, B, C, and D (electric hexadecapole interaction) in fitting, we have found it meaningless to include the D parameter because the deduced D parameter has the same order of magnitude of uncertainties. As was pointed out by Childs [22], the fitting with the four parameters made no improvement in reproducing the experimental frequencies.

B. Isotopic ratio of the hfs constants A, B, and C for ^{177}Hf and ^{179}Hf

From the corrected hfs constants A, B, and C, the isotopic ratios for ^{177}Hf and ^{179}Hf were obtained as summarized in Table III. A hfs anomaly that is well approximated to the relation [23]

$$^{177}\Delta^{179}=\frac{A(^{177}\mathrm{Hf})}{A(^{179}\mathrm{Hf})}\,\frac{g_I(^{179}\mathrm{Hf})}{g_I(^{177}\mathrm{Hf})}-1 \tag{4}$$

was also evaluated using the nuclear g factors g_I obtained by Büttgenbach *et al.* [5], and are listed in Table III. Although values of the hfs anomaly are hardly determined to be accurate enough to discuss in detail due to the large uncertainties of g_I [5], it is interesting to note that the ratio of $A(^{177}\mathrm{Hf})/A(^{179}\mathrm{Hf})$ has a significant J dependence, indicating J-dependent hyperfine anomalies. Since the hyperfine anomalies are considered to be important only in configurations involving unpaired s (or $p_{1/2}$) electrons, the observed J-dependent hyperfine anomalies are resulting from the different admixtures of $5d^3 6s$ configuration in the $5d^2 6s^2$ configuration, which are of an order of magnitude of 1% depending on J [24,25]. The J depen-

TABLE III. Isotopic ratios for the hfs constants A, B, and C, and hyperfine anomaly $^{177}\Delta^{179}$ for the $5d^2 6s^2\,{}^3F_J$ states in ^{177}Hf and ^{179}Hf. The numbers in the parentheses after A and B indicate the mass number.

State	$A(177)/A(179)$	$B(177)/B(179)$	$C(177)/C(179)$	$^{177}\Delta^{179}$
$^3F_2$	−1.588 060(1)	0.884 924(1)	−0.41(9)	−0.002 4(21)
$^3F_3$	−1.588 560(3)	0.884 929(3)	−0.38(22)	−0.002 1(21)
$^3F_4$	−1.588 814(5)	0.884 921(3)	−0.42(24)	−0.002 0(21)

dence of the ratio $A(^{177}\mathrm{Hf})/A(^{179}\mathrm{Hf})$ has an order of magnitude of 0.03% (see Table III), and the J dependence of the admixture of $5d^36s$ has an order of magnitude of 0.3% [25]. Relatively speaking, this is consistent with the case of Ir [26]: The J dependence of the ratio $A(^{191}\mathrm{Ir})/A(^{193}\mathrm{Ir})$ in the ^4F term of $5d^76s^2$ has an order of magnitude of 0.3%, and the J dependence of the admixture of $5d^86s$ has an order of magnitude of several percent.

As is seen from Table III, the isotopic ratios for B and C are constant for the ^3F_J ($J=2$–4) states within uncertainties. This is in accord with the hfs measurements of ^{151,153}Eu by Childs [22]. It should be mentioned, however, that unlike the case of ^{151,153}Eu where the isotopic ratio of C is close to unity [22], the constant C has a strong J dependence for both ^{177}Hf and ^{179}Hf (see Table II): The absolute values of the constants C are likely to increase with J for both ^{177}Hf and ^{179}Hf and they have opposite signs, corresponding to the signs of the constants A.

The isotopic ratio of C for ^{177}Hf and ^{179}Hf is probably taken as the corresponding isotopic ratio of the nuclear ground-state magnetic octupole moments Ω [22], i.e.,

$$\frac{\Omega(^{177}\mathrm{Hf})}{\Omega(^{179}\mathrm{Hf})}=\frac{C(^{177}\mathrm{Hf})}{C(^{179}\mathrm{Hf})}=-0.41(9)\ . \qquad (5)$$

Here we adopt the ratio for $^3F_2$ because experimental errors for $^3F_3$ and $^3F_4$ are too large. Evaluation of Ω for either ^{177}Hf or ^{179}Hf alone is very difficult at present, since we need to know the effective value of $\langle r^{-5}\rangle_{5d}$, which is likely to be strongly perturbed by octupole shielding or antishielding analogous to the well-known Sternheimer shielding for the quadrupole interaction [22].

The present investigation provides a clear indication of the nuclear-spin dependence of the magnetic octupole hfs constant C along with the case of ^{175,176}Lu [27]. Table IV summarizes the known octupole hfs constants and their isotopic ratios together with the data on Lu and Hf for comparison. As is seen from the table, the isotopic ratios for relatively large mass atoms are rather close to unity, except for Lu and Hf whose nuclear spins are different between their isotopes, respectively. It seems hard to explain the nuclear-spin dependence of the magnetic octupole hfs constants observed for Lu and Hf in terms of the nuclear single-particle model [28].

C. The Sternheimer shielding effect

To estimate the Sternheimer shielding effect for the electric quadrupole interaction [2,3], the single-electron hfs parameters $a_{5d}^{k_sk_l}$ and $b_{5d}^{k_sk_l}$ for the $5d^26s^2$ configuration were deduced for ^{177,179}Hf using Eqs. (2) and (3) from the corrected hfs constants A and B of the states ^3F_J ($J=2$–4) listed in Table II. For simplicity, the angular coefficients $\alpha_{5d}^{k_sk_l}$ and $\beta_{5d}^{k_sk_l}$ were calculated using the intermediate coupling wave functions given by Büttgenbach Dicke, and Gebauer [6]. Final values of $a_{5d}^{k_sk_l}$ and $b_{5d}^{k_sk_l}$ for the $5d^26s^2$ configuration are presented in Table V for ^{177,179}Hf, and the previous values [6] for ^{179}Hf are included for comparison.

By neglecting the configuration-interaction effects (configuration mixings), the relativistic parameters $a_{5d}^{k_sk_l}$ and $b_{5d}^{k_sk_l}$ can be related to the nonrelativistic parameters a_{5d} and b_{5d} [21], respectively. According to Büttgenbach and Meisel [4], $\alpha_{5d}^{k_sk_l}(b_{5d}^{k_sk_l})$ are related to $a_{5d}(b_{5d})$ as follows:

TABLE IV. Known magnetic octupole hfs constants C and their isotopic ratios together with their nuclear information.

	Nuclear configuration		Nuclear	C		
Isotope	Proton	Neutron	spin and parity	(Hz)	Ratio of C	Ref.
^{35}Cl	$d_{3/2}$		$\frac{3}{2}^+$	$-6.9(12)$	1.28(36)	[28]
^{37}Cl	$d_{3/2}$		$\frac{3}{2}^+$	$-5.4(12)$		
^{69}Ga	$p_{3/2}$		$\frac{3}{2}^-$	84(6)	0.73(7)	[29]
^{71}Ga	$p_{3/2}$		$\frac{3}{2}^-$	115(7)		
^{79}Br	$p_{3/2}$		$\frac{3}{2}^-$	388(8)	0.90(3)	[30]
^{81}Br	$p_{3/2}$		$\frac{3}{2}^-$	430(8)		
^{113}In	$g_{9/2}$		$\frac{9}{2}^+$	1728(45)	1.02(3)	[31]
^{115}In	$g_{9/2}$		$\frac{9}{2}^+$	1702(35)		
^{151}Eu	$d_{5/2}$		$\frac{5}{2}^+$		0.87(6)	[22]
^{153}Eu	$d_{5/2}$		$\frac{5}{2}^+$			
^{175}Lu	$g_{7/2}$		$\frac{7}{2}^+$	$-135(23)$	0.21(7)	[27]
^{176}Lu	$g_{7/2}$	$f_{7/2}$	7^-	$-654(175)$		
^{177}Hf		$f_{7/2}$	$\frac{7}{2}^-$	0.17(3)	$-0.41(9)$	Present
^{179}Hf		$i_{13/2}$	$\frac{9}{2}^+$	$-0.41(5)$		

TABLE V. Single-electron hfs parameters $a_{5d}^{k_s k_l}$ and $b_{5d}^{k_s k_l}$ in MHz obtained from hfs constants A and B for the $5d^2 6s^2$ configuration. Previous values for ^{179}Hf (Ref. [6]) are given for comparison in the bottom row.

Isotope	a_{5d}^{01}	a_{5d}^{12}	a_{5d}^{10}	b_{5d}^{02}	b_{5d}^{13}	b_{5d}^{11}
^{177}Hf	85.7053(2)	23.2453(15)	15.7596(7)	4662.694(5)	2324.645(25)	−882.206(9)
^{179}Hf	−53.9503(3)	−14.7006(26)	−9.9052(12)	5269.049(9)	2627.073(53)	−996.966(17)
	−53.95	−14.68	−9.90	5269.04	2626.94	−996.90

$$a_{5d}^{01}=1.0556a_{5d},\quad a_{5d}^{12}=1.1401a_{5d},\quad a_{5d}^{10}=-0.0273a_{5d}\ ; \tag{6}$$

$$b_{5d}^{02}=1.1111b_{5d},\quad b_{5d}^{13}=0.4444b_{5d},\quad b_{5d}^{11}=-0.076b_{5d}\ .$$

The ratios of a_{5d}^{12}/a_{5d}^{01}, a_{5d}^{10}/a_{5d}^{01}, b_{5d}^{13}/b_{5d}^{02}, and b_{5d}^{11}/b_{5d}^{02} obtained from the above relations considerably deviate from the present experimental values, particularly for a_{5d}^{10}/a_{5d}^{01} (see Table V). This fact also shows that the configuration-interaction effects (configuration mixings) are striking for the $5d^2 6s^2$ configuration in question.

Since they are less sensitive to the configuration-interaction effects [21], the parameters a_{5d}^{01} and b_{5d}^{02} were used to derive the nonrelativistic parameters a_{5d} and b_{5d}. The electric quadrupole moment Q'_{5d} including the Sternheimer shielding effect [2,3], is written as

$$Q'_{5d}=\frac{2\mu_B\mu_N}{e^2}\frac{\mu_I}{I}\frac{b_{5d}}{a_{5d}}\ , \tag{7}$$

where μ_B and μ_N are the Bohr magneton and the nuclear magneton, respectively, and μ_I is the nuclear magnetic dipole moment [21]. The spectroscopic quadrupole moment Q is related to Q'_{5d} as

$$Q=Q'_{5d}/(1-R_{5d})\ , \tag{8}$$

where R_{5d} is the Sternheimer shielding factor taking into account the influence of the orbital polarization of the electron core on the nuclear quadrupole measurements from hfs.

The derived values of Q'_{5d} for ^{177,179}Hf are

$$Q'_{5d}(^{177}\text{Hf})=4.699(4)\text{b}\ ,$$

$$Q'_{5d}(^{179}\text{Hf})=5.299(11)\text{b}\ .$$

Using the known spectroscopic quadrupole moments Q from the muonic x-ray measurements [13], the Sternheimer shielding factors R_{5d} are taken to be

$$R_{5d}(^{177}\text{Hf})=-0.396(12)\ ,$$

$$R_{5d}(^{179}\text{Hf})=-0.397(12)\ .$$

These values should be considered the same within uncertainties. This is confirmed by the fact that the shielding factor should be independent of isotopes. We have an average value of $R_{5d}=-0.397(8)$, which is considered to be in agreement with the most probable value obtained for ^{179}Hf by Büttgenbach, Dicke, and Gebauer [6] and those for ^{191,193}Ir by Tanaka *et al.* [14]. The values R_{5d} for ^{177,179}Hf by Tanaka *et al.* [14], which are unreasonably large compared to those for ^{191,193}Ir and rare-earth elements [14], are not acceptable because they used an old estimation of Q' by Büttgenbach *et al.* [5], where only the ground state $^3F_2$ was taken into account and the configuration mixing effect was neglected, i.e., the use of Eq. (6). As is seen from Table V, the revised data on the single-electron hfs parameters for ^{179}Hf by Büttgenbach, Dicke, and Gebauer [6], where they measured the hfs of $^3F_3$, $^3F_4$ as well as $^3F_2$, are in good agreement with the present data. This means that the effect of the configuration mixing is important in deducing the Sternheimer shielding factor for the 5*d* elements.

The spectroscopic quadrupole moments Q of ^{177,179}Hf reported by Büttgenbach *et al.* [5] deviate largely from the values determined from muonic x-ray measurements [13]. Now it should be pointed out that the main source of such a deviation is in the use of inappropriate single-electron hfs parameters, since the Sternheimer shielding factor $R_{5d}=-0.3$ was used in their analysis.

V. SUMMARY

The hfs splittings in the $5d^2 6s^2\,{}^3F_J$ ($J=2$–4) states in ^{177,179}Hf I have been precisely measured by means of the atomic-beam laser-rf double-resonance technique. This is a measurement of LRDR in Hf I. Second-order hfs corrections have been found to be important in determining the magnetic dipole (A), electric quadrupole (B) and, particularly, magnetic octupole (C) hfs constants with high accuracy. After corrections for the second-order hfs effects, the hfs constants A, B, and C have been obtained for the $5d^2 6s^2\,{}^3F_J$ ($J=2$–4) states in ^{177,179}Hf I.

J dependences of the isotopic ratio of the hfs constants A for ^{177}Hf and ^{179}Hf show J-dependent hyperfine anomalies, resulting from the configuration mixing of $5d^3 6s$ in an order of magnitude of 1%. The hfs constant C shows a remarkable J dependence as well as does constants A and B. The isotopic ratio of C is, however, constant within experimental uncertainties, which is taken to be the ratio of the nuclear-ground-state magnetic octupole moments Ω, i.e., $\Omega(^{177}\text{Hf})/\Omega(^{179}\text{Hf})=-0.41(9)$. The magnetic octupole hyperfine interaction has been found to be strongly dependent on the nuclear spin.

Single-electron hfs parameters $a_{5d}^{k_s k_l}$ and $b_{5d}^{k_s k_l}$ of the 5*d* electron have been obtained and the configuration-interaction effects have been found to be significantly strong for the $5d^2 6s^2$ configuration in Hf I. The Sternheimer shielding factor is derived to be $R_{5d}=-0.397(8)$.

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