Isotope shift and hyperfine structure in Lu I and W I

W. G. Jin, M. Wakasugi, and T. T. Inamura*
Cyclotron Laboratory, The Institute of Physical and Chemical Research (RIKEN), Wako, Saitama 351-01, Japan

T. Murayama
Tokyo University of Mercantile Marine, Koto, Tokyo 135, Japan

T. Wakui and H. Katsuragawa
Department of Physics, Toho University, Funabashi, Chiba 274, Japan

T. Ariga and T. Ishizuka
Department of Physics, Saitama University, Urawa, Saitama 338, Japan

M. Koizumi
Japan Atomic Energy Research Institute, Takasaki, Gunma 370-12, Japan

I. Sugai
Insitute for Nuclear Study, University of Tokyo, Tanashi, Tokyo 188, Japan
(Received 10 September 1993)

Isotope shifts and hyperfine structures of four transitions in W I have been measured as well as isotope shifts in Lu I by means of atomic-beam laser spectroscopy. Magnetic-dipole hyperfine constants A of ^{183}W are determined for the atomic states $^7D_1$, $^7F_1$, and $^7F_3$ of the $5d^46s6p$ configuration. The nuclear parameters λ and $\delta\langle r^2\rangle$ are derived for W stable isotopes. J dependences of the isotope shifts are observed for the ^2D term of the $5d6s^2$ configuration in Lu I and the ^5D term of the $5d^46s^2$ configuration in W I. Parameters z_{5d} of the crossed-second-order effects are derived for the Lu I $5d6s^2$ and W I $5d^46s^2$ configurations, and z_{5d}/λ is found to be 411(26) MHz/fm^2 for W. A systematic behavior of the normalized parameter $z_{nl}/\lambda\zeta_{nl}$ is discussed.

PACS number(s): 32.30.Jc, 32.70.Jz, 35.10.Fk, 21.90.+f

I. INTRODUCTION

Measurements of isotope shift (IS) and hyperfine structure (hfs) by means of laser spectroscopy have been intensively carried out over the last decade and yielded important information on nuclear structures as well as electronic configurations, electron densities, and atomic structures [1–6]. Recently, considerable attention has been paid to the crossed-second-order (CSO) effect associated with the term and J dependences of IS for the $5d$ and $4f$ elements [7–9]. No systematic behavior, however, has been found and the CSO effect is far from being well understood.

For the isotopes of ^{175,176}Lu, which are of a $5d$ element, investigators reported measurements of IS and hfs [10–15]. The hf constants A and B were measured for the ground-state term ^2D of the $5d6s^2$ configuration precisely by the atomic-beam magnetic resonance [10] and the ^4F term and the $^2D_{3/2}$ state of the $5d6s6p$ configuration by laser spectroscopy [11–15]. Isotope shifts of $^{175-176}$Lu were also measured for the three transitions from the ground-state term ^2D of the $5d6s^2$ configuration to the upper term ^4F of the $5d6s6p$ configuration [11–13]. However, no report has been made for J dependence of IS in Lu I.

For ^{183}W, which is another $5d$ element, the magnetic hf constants A have been precisely determined for the ground-state term ^5D of the $5d^46s^2$ configuration and the $^7S_3$ state of the $5d^56s$ configuration by means of the atomic-beam magnetic resonance [16,17]. For the excited states, the hf constants A for the levels at 20 064 and 26 367 cm^{-1} have been measured by means of the Fabry-Pérot spectrometer [18]. As for the IS of W, several measurements were previously made by means of the conventional Fabry-Pérot spectrometer with the hollow cathode [18–22], but no accurate measurement like laser spectroscopy has so far been reported. Although Aufmuth, Steudel, and Wöbker [21] reported a large term dependence of IS for the terms ^7S and ^5S of the $5d^56s$ configuration in W I and derived parameters z_{5d} of the CSO effect for the $5d^46s^2$ configuration in W I, their values of z_{5d} are unreasonably large compared with those of neighboring elements.

Therefore it is most desirable to carry out laser spectroscopic measurement of hfs and IS for Lu and W isotopes to study J dependence of IS in $5d$ elements systematically. We shall report laser spectroscopy of stable

*Present address: Safety Center, The Insitute of Physical and Chemical Research (RIKEN), Wako, Saitama 351-01, Japan.

isotopes of W, along with laser spectroscopy of ^{175,176}Lu, and discuss J dependence of IS in detail. Our newly developed Ar-ion-sputtering atomic-beam source has enabled this kind of work on refractory elements to be easily made [23].

II. EXPERIMENTAL METHOD

The present experiment was carried out by means of laser spectroscopy with the Ar-ion-sputtering atomic-beam source [23]. The Ar-ion beam generated by an electron gun was accelerated to 8 keV and focused to about 1 mm^2 on the Lu or W natural metallic target: the isotopic compositions are 97.39% ^{175}Lu and 2.61% ^{176}Lu for the Lu target; 28.6% ^{186}W, 30.7% ^{184}W, 14.3% ^{183}W, 26.3% ^{182}W, and 0.13% ^{180}W for the W target. Atoms of up to 10^{11} per second were obtained in the laser–atomic-beam interacting region.

A cw ring dye laser (Coherent 699-29) pumped with an Ar-ion laser (Spectra Physics 171-19) was operated using dyes of rhodamine 6G and rhodamine 110 to cover a wavelength range of 530–620 nm. To reduce the Doppler broadening, both the atomic beam and the laser beam were collimated and the laser beam crossed the atomic beam perpendicularly. Laser-induced fluorescent lights focused by a spherical mirror were detected with a cooled photon-counting photomultiplier (Hamamatsu R1333) and recorded in a computer. To make accurate determination of peak separations, a confocal Fabry-Pérot interferometer (FPI) with a free spectral range of 150 MHz was used to produce a set of frequency markers recorded simultaneously with the fluorescence spectra from the atomic beam. The experimental setup has been described in detail previously [23].

III. EXPERIMENTAL RESULTS

Four optical transitions studied for Lu I and W I and related energy level schemes [24,25] are shown in Fig. 1. Transitions in Lu I are from the ground state $^2D_{3/2}$ and the lower metastable state $^2D_{5/2}$ of the $5d6s^2$ configuration and transitions in W I are from the lower metastable states of the ground-state term ^5D of the $5d^46s^2$ configuration.

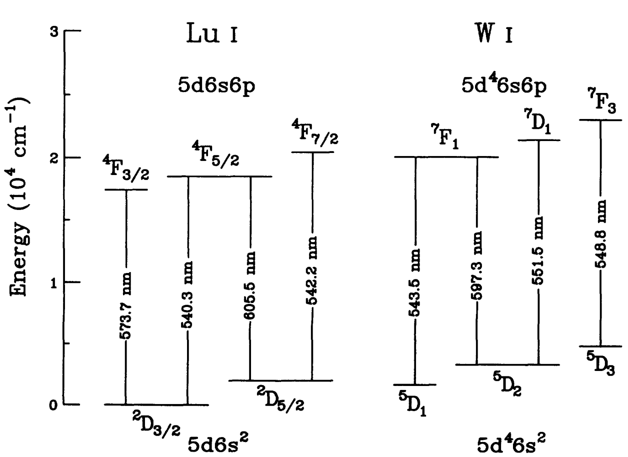

FIG. 1. Transitions of interest and related energy level schemes in Lu I and W I.

Typical fluorescence spectra of 540.3-nm transition in Lu I and 597.3-nm transition in W I are shown in Figs. 2 and 3, respectively. It is seen from Fig. 2 that the 12 hf peaks of ^{175}Lu are all observed while there hf peaks of ^{176}Lu are overlapped with the large hf peaks of ^{175}Lu. It is seen from Fig. 3 that peaks of both the even-A and odd-A W isotopes have been observed being well separated; even for the isotope ^{180}W with the smallest abundance of 0.13% the peak has been identified. Three hf peaks of the odd-A isotope ^{183}W are clearly seen. The full width at half maximum (FWHM) of the peak was about 45 MHz for Lu and W, which is mainly resulting from the velocity distribution of the sputtered atomic beam [23]. The background in the spectra is mainly from the sputtered atomic beam itself.

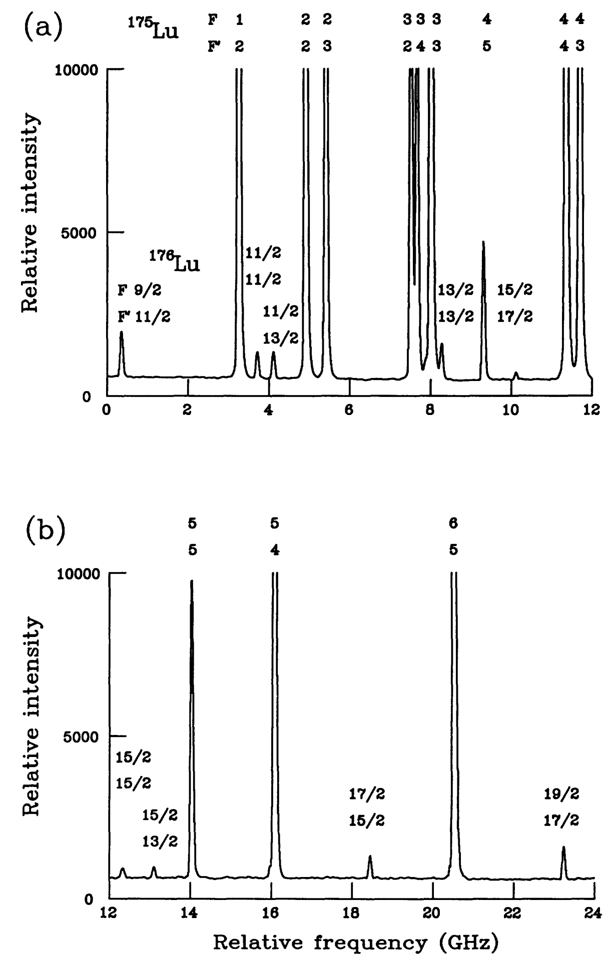

FIG. 2. Measured hf spectrum of the 540.3-nm transition in Lu I. The spectrum (a) shows the lower half of the observed spectrum as a function of laser frequency, and (b) the upper half. Strong peaks are the hf peaks of ^{175}Lu and weak ones those of ^{176}Lu. Both peaks of ^{175}Lu and ^{176}Lu are labeled with a pair of the total angular momenta F and F': F for the upper state; F' for the lower state.

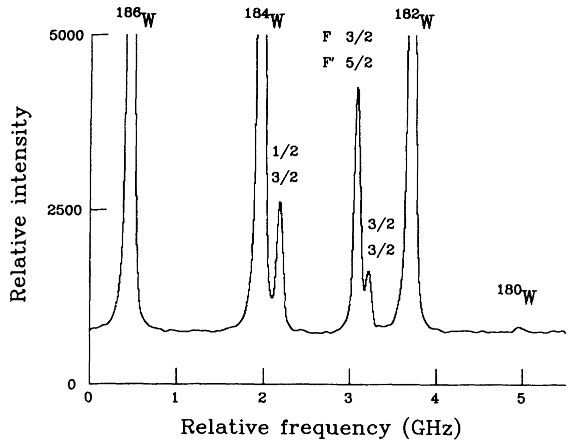

FIG. 3. Measured spectrum of the 597.3-nm transition in W I. Peaks of even-A isotopes are labeled with isotopic symbols. The hf peaks of the odd-A isotope ^{183}W are labeled with a pair of the total angular momenta F and F' as in Fig. 2.

A. Hyperfine interaction

Since the hyperfine structures of ^{175,176}Lu are known for the ground-state term ^2D of the $5d6s^2$ configuration and the ^4F term of the $5d6s6p$ configuration [10–13], here we shall discuss the hyperfine structures of ^{183}W only.

The atomic nucleus ^{183}W possesses a nuclear spin $I=\frac{1}{2}$ in the ground state. Since the hf interaction for such an atomic nucleus is given by the magnetic-dipole interaction only [26], the hfs separation $\Delta\nu$ between $F=J+\frac{1}{2}$ and $J-\frac{1}{2}$ is related to the magnetic-dipole hf constant A by

$$\Delta\nu = A(J+\tfrac{1}{2}) , \qquad (1)$$

where J is the electronic angular momentum and F is the total angular momentum of the atom.

The hfs separation $\Delta\nu$ and hyperfine constants A thus determined are summarized in Table I for the states $^7D_1$, $^7F_1$, and $^7F_3$ of the $5d^46s6p$ configuration in ^{183}W I. The previous value A for the $^7F_1$ state at 20 064 cm^{-1} is included for comparison [18]. It is seen that the present values A of the $5d^46s6p$ configuration are very large compared with those of the ground-state $5d^46s^2$ configuration [$A(^5D_J)\approx 29-88$ MHz] [16,17]. This is reasonable because the open-shell $6s$ and $6p$ electrons of the $5d^46s6p$ configuration have much greater contributions to the constant A, i.e., the atomic magnetic field, than those of the $5d^46s^2$ configuration.

TABLE I. Hyperfine structure separations $\Delta\nu$ and magnetic-dipole hyperfine constants A determined for ^{183}W.

State	Energy (cm^{-1})	$\Delta\nu$ (MHz)	A (MHz) Present	A (MHz) Previous[a]
$5d^46s6p\ ^7F_1$	20 064.30	1098.1(24)	732.1(16)	729(9)
$5d^46s6p\ ^7D_1$	21 453.90	1624.7(15)	1083.1(10)	
$5d^46s6p\ ^7F_3$	23 047.31	655.9(10)	437.27(67)	

[a]Reference [18].

TABLE II. Measured isotope shifts of $^{176-175}$Lu for four transitions in Lu I.

Transition (nm)	Isotope shift (MHz)
573.7	−394.1(13)
540.3	−388.8(11)
605.5	−418.0(12)
542.2	−409.2(12)

B. Isotope shift

The obtained isotope shifts for four transitions studied (see Fig. 1) are listed in Tables II and III for Lu and W, respectively. The isotope shifts for the even-A W isotopes were obtained from the relative frequencies of the peaks in the measured spectra and those of the isotopes ^{175,176}Lu and ^{183}W were deduced from the center of gravity of the hf splitting. For the 548.8-nm transition in W I, the peak of ^{180}W was not observed because of its weak intensity. The uncertainty of IS consists of three parts: (1) the uncertainty of the peak center, (2) the uncertainty from the frequency calibration of FP spectra, and (3) the systematic error of the free spectral range of the FPI (0.02%). To reduce the uncertainty of IS to less than 0.3%, measurements were repeated more than five times.

IV. ANALYSIS AND DISCUSSION

A. Isotope shift

The observed isotope shift $\delta\nu_i$, between isotopes with mass number A and A' for a transition i, consists of the normal mass shift (NMS), the specific mass shift (SMS), and the field shift (FS). The NMS is calculated easily [27] while it is very difficult to calculate the SMS. The FS $\delta\nu_{i\,\mathrm{FS}}$ is given in the first-order perturbation theory (for instance, see Ref. [27]) by

$$\delta\nu_{i\,\mathrm{FS}} = E_i f(Z)\lambda , \qquad (2)$$

where E_i and $f(Z)$ are the electronic and relativistic correction factors, respectively. The nuclear parameter λ is related to the changes in mean-square nuclear charge radii $\delta\langle r^2\rangle$ and higher-order contributions [27]:

$$\lambda = \delta\langle r^2\rangle + \frac{C_2}{C_1}\delta\langle r^4\rangle + \frac{C_3}{C_1}\delta\langle r^6\rangle + \cdots , \qquad (3)$$

where the expansion coefficients C_1, C_2, and C_3 have been tabulated by Seltzer [28].

Figure 4 shows King plots for the transitions measured in W I. Coordinates are given by the modified isotope shifts $\delta\nu_i^{\mathrm{mod}}$ which are defined as [27,29]

$$\delta\nu_i^{\mathrm{mod}} = (\delta\nu_i - \delta\nu_{i\,\mathrm{NMS}})\frac{AA'}{A'-A}\,\frac{2}{186\times 184} . \qquad (4)$$

The ($5d^46s^2\ ^5D_1 - 5d^46s6p\ ^7F_1$) 543.5-nm transition is chosen as a reference, i.e., $\delta\nu_{543.5\ \mathrm{nm}}^{\mathrm{mod}}$ is on the abscissa. Since it is determined by the ratio E_i/E_j, the slope of the

TABLE III. Measured isotope shifts of stable isotopes for four transitions in W I.

Transition (nm)	Isotope shift (MHz) 186-184	184-182	183-182	182-180
543.5	−1582.7(24)	−1829.9(15)	−960.5(15)	−1274.2(19)
597.3	−1601.2(10)	−1850.1(16)	−971.3(12)	−1290.9(14)
551.5	−1384.2(14)	−1597.5(12)	−837.9(12)	−1118.1(21)
548.8	−1614.9(17)	−1862.0(14)	−977.9(13)	

King plot should be the same and equal to unity for the transitions of the same type: here the ns^2-$nsnp$ transitions. The slope of the King plot for the 551.5-nm transition, however, turned out to be 0.862(6), which is significantly smaller than unity; the other two transitions of 597.3 and 548.8 nm show their slopes equal to unity within experimental uncertainties. Such deviation from unity for the 551.5-nm transition should be attributed to the large term dependence of IS or the configuration mixing. According to Aufmuth *et al.* [22], the term dependence of IS should be very small for the ^7F and ^7D terms, so that strong configuration mixing should exist in the $^7D_1$ state of the $5d^46s6p$ configuration with which the 551.5-nm transition is associated.

To deduce the field shift from the observed IS, we assume that the 543.5-nm transition is a pure s^2-sp transition, adopting the semiempirical evaluation [27] of SMS=(0.0±0.5) NMS. Then we have the specific mass shifts and field shifts for the other transitions from the King plots, which are summarized in Table IV along with the calculated normal mass shifts. For the 551.5-nm transition, FS is about 200 MHz larger than those for the other transitions which have almost the same values of FS within uncertainties. This feature of the 551.5-nm transition is also understood as a result of the configuration mixing in the $^7D_1$ state in question although it is hard at present to describe quantitatively.

B. The nuclear parameter λ and $\delta\langle r^2\rangle$ of W

From the derived field shifts, the relative values λ were extracted [see Eq. (2)] for the W stable isotopes. To obtain the absolute value λ we have to know the electronic factor E_i and the relativistic correction factor $f(Z)$. The electronic factor E_i is derived to be 0.4089 for the $5d^46s^2$-$5d^46s6p$ transition according to Otten [1] using the hf parameter a_{6s} [17,30] and the screening factor γ [21]. The relativistic correction factor $f(Z)$ is calculated to be 48.55 GHz/fm^2 according to Ahmad *et al.* [31] using the isotope-shift constant $C_{\rm unif}$ tabulated for the nucleus of a uniformly charged sphere [32]. The theoretical uncertainty of 5% (for instance, see Ref. [33]), which results from the uncertainties of E_i and $C_{\rm unif}$, is taken into account in calculating $E_if(Z)$. Relative and absolute values of λ thus obtained are listed in Table V together with previous values compiled by Aufmuth, Heilig, and Steudel [34] for comparison. It is seen from Table V that the values λ from the present laser spectroscopic measurement agree with those of the previous measurements [34] which were made using the FP spectrometer [18–22].

To deduce $\delta\langle r^2\rangle$ from the values λ, the two-parameter model [1,31] was used. The obtained values $\delta\langle r^2\rangle$ for the W stable isotopes are also presented in Table V. No values of $\delta\langle r^2\rangle$ have previously been reported on W; Aufmuth, Heilig, and Steudel [34] presented the values λ only, and Heilig and Steudel [27] simply assumed $\lambda=\delta\langle r^2\rangle$.

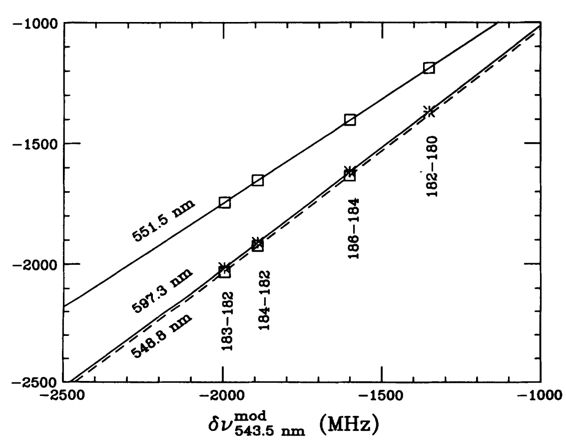

FIG. 4. King plots of the isotope shifts for the $5d^46s^2$-$5d^46s6p$ transitions in W I. Straight lines are the least-squares fits; experimental uncertainties are within symbols.

C. *J* dependence of the isotope shift in the Lu I $5d6s^2$, $5d6s6p$, and W I $5d^46s^2$ configurations

The 540.3 and 605.5-nm transitions in Lu I belong to the same upper level $^4F_{5/2}$ (see Fig. 1). Therefore the IS difference between the two transitions should be that between the lower levels of $^2D_{3/2}$ and $^2D_{5/2}$, i.e., a residual level isotope shift $T_{\rm re}$ [3,35], which is a measure of *J* dependence of IS within a term. Residual isotope shifts $T_{\rm re}$ were thus obtained for $^2D_{5/2}$ with respect to $^2D_{3/2}$ of the $5d6s^2$ configuration in Lu I and also $^4F_{5/2}$, $^4F_{7/2}$ with respect to $^4F_{3/2}$ of the $5d6s6p$ configuration in Lu I, and are presented in Table VI. Similarly, residual isotope shifts $T_{\rm re}$ were obtained for $^5D_2$ with respect to $^5D_1$ of the $5d^46s^2$ configuration in W I as presented in Table VII.

The *J* dependence of IS results from the crossed-second-order effect or the far-configuration-mixing effect [7]. The CSO contribution to the isotope shift of a pure configuration state with the zeroth-order wave function ψ_0 and the energy E_0 is written as [7]

$$E_{\rm CSO}=2\sum_x \frac{\langle\psi_0|\hat{Q}|\psi_x\rangle\langle\psi_x|\hat{O}|\psi_0\rangle}{E_0-E_x}\ , \tag{5}$$

TABLE IV. Normal mass shifts (NMS), specific mass shifts (SMS), and field shifts (FS) (all in MHz) of stable isotopes for four transitions in W I.

Transition (nm)		543.5	597.3	551.5	548.8
NMS	186-184	19.2	17.5	18.9	19.0
	184-182	18.7	17.1	18.5	18.6
	183-182	9.3	8.5	9.2	9.2
	182-180	18.3	16.7	18.1	
SMS	186-184	0(9)	−7(13)	−23(13)	−22(22)
	184-182	0(9)	−7(13)	−22(13)	−22(22)
	183-182	0(5)	−3(7)	−11(6)	−11(11)
	182-180	0(9)	−7(13)	−22(13)	
FS	186-184	−1602(10)	−1612(13)	−1381(13)	−1612(22)
	184-182	−1849(9)	−1861(13)	−1594(13)	−1859(22)
	183-182	−970(5)	−976(7)	−836(7)	−976(11)
	182-180	−1292(9)	−1301(13)	−1115(13)	

where ψ_x and E_x are the zeroth-order wave functions and the energies of other configurations, respectively. $\hat{Q}$ represents the operator of the electrostatic interaction or the magnetic interaction of all electrons; $\hat{O}$ represents the IS operator. The sum ranges over all states x of all other electronic configurations. The CSO effect between the electrostatic interaction and IS operators leads to the term dependence of IS in a pure configuration, whereas the CSO effect between the magnetic interaction and IS operators leads to the J dependence of IS in a term of the pure configuration.

The J dependence of IS is described by one parameter z_{nl} with the angular coefficient c_{nl} that is the same as the one entering into the spin-orbit interaction energy [7]. Accordingly, the residual IS $T_{\rm re}$ within a term for the $5d^N6s^2$ configuration can be written as

$$T_{\rm re}=\Delta c_{5d}z_{5d} \ . \tag{6}$$

For the $5d6s^2$ configuration in Lu I, the ^2D_J state should be a pure LS coupling state consisting of the $5d$ electron only; and then we have $\Delta c_{5d}=2.5$. The parameter z_{5d} thus obtained for the $5d6s^2$ configuration in Lu I is presented in Table VI. For the $5d6s6p$ configuration in Lu I, configuration mixing and intermediate wave functions should be considered and CSO contributions from the $6p$ electron should be included. Because no information about the wave functions is available for the ^4F_J state, it is impossible to make further analysis for the $5d6s6p$ configuration in Lu I.

For the $5d^46s^2$ configuration in W I, since the intermediate wave function is not known for the ^5D_J level, we have calculated Δc_{5d}, assuming the ^5D_J level to be a pure LS coupling state. The values z_{5d} derived for different isotope pairs are listed in Table VII; those obtained by Aufmuth, Steudel, and Wöbker [21] are also included in Table VII for the isotope pairs of 186-184 and 186-182. The latter values are about 3.5 times larger than the present values. It should be pointed out, however, that Aufmuth, Steudel, and Wöbker deduced their values of z_{5d} from the six levels belonging to different terms of the $5d^46s^2$ and $5d^56s$ configurations, where a fit procedure involving large term dependences for ^7S and ^5S had to be used [21], as they mentioned, their determination by means of the fit procedure is subject to large uncertainties since large alterations of z_{5d} have almost no influence on the results of the fit. In contrast, we have obtained the present values z_{5d} directly from the ground-state term ^5D of the $5d^46s^2$ configuration in W I without any uncertainty inherent in the fit procedure.

To see contributions of the field shift and the specific mass shift in the CSO effect, we divide the values z_{5d} of the $5d^46s^2$ configuration in W I by the nuclear parameter λ; the results are listed in Table VII. It is seen that the values z_{5d}/λ are constant for different isotope pairs within uncertainties. This indicates that the field shift has dominant contribution and the SMS is negligible in the CSO effect. An average value of z_{5d}/λ is obtained to be 411(26) MHz/fm^2.

TABLE V. Derived relative and absolute values of nuclear parameters λ and changes in mean-square nuclear charge radii $\delta\langle r^2\rangle$ for W.

Isotope pair	Relative λ Present	Relative λ Previous[a]	Absolute λ (fm^2) Present	Absolute λ (fm^2) Previous[a]	$\delta\langle r^2\rangle$ (fm^2)
186-184	1	1	0.0807(41)	0.084(7)	0.0852(44)
184-182	1.1542(40)	1.154(4)	0.0931(47)	0.097(8)	0.0986(51)
183-182	0.6056(21)	0.607(5)	0.0489(25)	0.051(5)	0.0518(27)
182-180	0.8071(42)	0.808(23)	0.0651(33)	0.068(8)	0.0683(35)

[a]Reference [34].

TABLE VI. Residual isotope shifts T_{re} and CSO parameters z_{5d} for the $5d6s^2$ and $5d6s6p$ configurations in Lu I.

Configuration	Level	T_{re} (MHz)	z_{5d} (MHz)
$5d6s^2$	$^2D_{3/2}$	0	
	$^2D_{5/2}$	28.2(16)	11.28(64)
$5d6s6p$	$^4F_{3/2}$	0	
	$^4F_{5/2}$	4.7(17)	
	$^4F_{7/2}$	12.5(24)	

D. Systematic behavior of the normalized parameter $z_{nl}/\lambda\zeta_{nl}$

Recently, attempts have been made to see the general behavior of the CSO parameters for the $4f$ and $5d$ elements [8,9]. Kronfeldt, Ashkenasi, and Nikseresht [8] reported that values of z_{4f}/λ had the same trend as the spin-orbit radial integral ζ_{4f}, and that those of $z_{4f}/\lambda\zeta_{4f}$ lay in the same order of magnitude for the $4f$ elements and increased slightly with increasing nuclear mass number. Sawatzky and Winkler [9] compared values of z_{5d}/λ for the $5d$ elements and pointed out that the scattering of the values z/λ apparently needs further consideration. Using the presently observed values for Lu and W and the previous values for other elements, we have noted an interesting systematic behavior of the CSO parameter.

The CSO parameter z_{nl} consist of the spin-orbit (magnetic) interaction and the IS (FS and SMS) effect [see Eq. (5)]. The spin-orbit interaction is expressed by the spin-orbit radial integral ζ_{nl} and the FS contains a nuclear part expressed by the nuclear parameter λ. Since the SMS is negligibly small in the CSO effect, z_{nl} is expressed as

$$z_{nl} = z_{nl\,\mathrm{norm}}\lambda\zeta_{nl} , \quad (7)$$

where the normalized parameter $z_{nl\,\mathrm{norm}}$ should be independent of the nuclear part and the spin-orbit (magnetic) interaction, and only depends on the electronic wave function in the CSO effects [8]. Here we shall discuss the systematic behavior of this normalized parameter $z_{nl}/\lambda\zeta_{nl}$.

For the $5d$ elements, the values z_{5d} in the $(5d+6s)^N$ ground-state configuration have already been reported for Hf, Re, Os, Ir, and Pt [9,36–39]. Using the values λ given by Aufmuth, Heilig, and Steudel [34] for Hf, Re, Os, Ir, and Pt, and values ζ_{5d} presented by Büttgenbach [30], the values $z_{5d\,\mathrm{norm}}$ are obtained and the results are shown in Fig. 5. The data reported by Aufmuth, Steudel, and Wöbker [21] for W are also shown in Fig. 5 for comparison. By neglecting the data point of Aufmuth, Steudel and Wöbker for W, it is seen from Fig. 5 that $z_{5d\,\mathrm{norm}}$ changes systematically as a function of the number of electrons N in the ground-state configuration in such a way that it increases with decreasing the number of electrons (or holes) occupying the shell of the ground-state configuration.

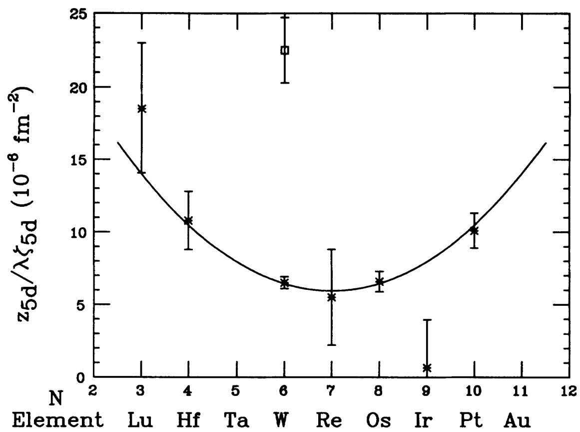

FIG. 5. Normalized parameters $z_{5d}/\lambda\zeta_{5d}$ for $5d$ elements in the $(5d+6s)^N$ ground-state configuration. N is the number of electrons in the configuration. The solid line is the least-squares fit of a quadratic function to the data; the data on W obtained by Aufmuth, Steudel, and Wöbker [21] (open square □) were neglected in the fit.

We have made a least-squares fit to the data with a quadratic function $a(N-b)^2+c$, which is shown with a solid curve in Fig. 5: $a=0.50(12)$, $b=7$, and $c=5.94(41)$. The parameter b corresponds to the number of electrons in the midshell with the orbital angular momentum l, i.e., $b=(2l+1)+2$, where an additional number 2 is the occupation number of the s shell; and $(N-b)^2$ is a measurer of the degree of freedom of electrons. Here the degree of freedom may be defined by $(N-b)^2/(1-b)^2$ with respect to a single electron (or hole). Such a systematical behavior of $z_{nl\,\mathrm{norm}}$ is also found for the $4f$-shell elements, as shown in Fig. 6, when the normalized parameters $z_{4f}/\lambda\zeta_{4f}$ [8,40] are plotted for the $(4f+5d+6s)^N$ ground-state configurations. The

TABLE VII. Residual isotope shifts T_{re} of the $^5D_2$ state and CSO parameters z_{5d} for the $5d^46s^2$ configuration in W I.

Isotope pair	T_{re} (MHz) $^5D_1$	$^5D_2$	z_{5d} (MHz) Present	Aufmuth, Steudel, and Wöbker[a]	z_{5d}/λ (MHz/fm^2)
186-184	0	16.7(26)	33.4(52)	114(18)	414(67)
186-183	0	25.2(32)	50.4(63)		403(55)
186-182	0	35.2(39)	70.3(77)	249(24)	405(49)
186-180	0	50.3(46)	100.6(93)		421(44)

[a]Reference [21].

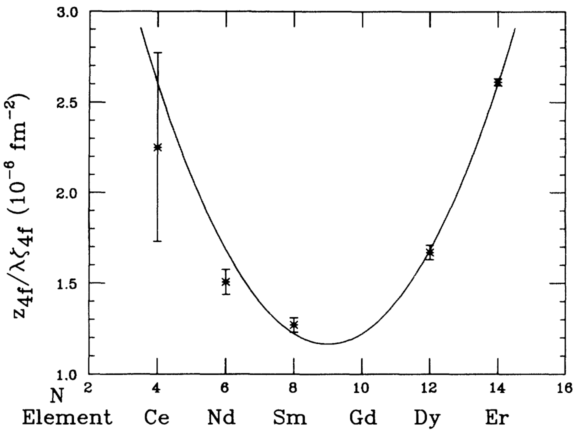

FIG. 6. Normalized parameters $z_{4f}/\lambda\zeta_{4f}$ for the rare-earth elements in the $(4f+5d+6s)^N$ ground-state configuration. The solid line is the least-squares fit of a quadratic function to the data.

solid curve is a quadratic fit to the data: $a=0.057(2)$, $b=9$, and $c=1.16(3)$. We conclude that $z_{nl\,\mathrm{norm}}$, i.e,. the CSO effect, is proportional to the degree of freedom of electrons (or holes) in the ground-state configuration.

It should be noted that the absolute value of $z_{4f\,\mathrm{norm}}$ is one order of magnitude smaller than that of $z_{5d\,\mathrm{norm}}$. This is considered to be due to the fact that the f shell is farther from the nucleus than the d shell and that interaction between electrons and the nucleus is much weaker for the f shell than for the d shell.

V. SUMMARY

We have succeeded in making laser spectroscopic measurement of W I and Lu I. The hf constants A of ^{183}W have been determined for the three states $^7F_1$, $^7D_1$, and $^7F_3$. The relative and absolute values of λ and the values of $\delta\langle r^2\rangle$ have been derived for the W stable isotopes, and J dependences of IS clearly seen for the ^2D term of the $5d6s^2$ configuration in Lu I and the ^5D term of the $5d^46s^2$ configuration in W I. The CSO parameters z_{5d} have been derived for the Lu I $5d6s^2$ and the W I $5d^46s^2$ configurations. The quantity of z_{5d}/λ is found to be constant for W isotopes, being 411(26) MHz/fm^2. This means that the field shift has dominant contribution in the CSO effect. It is found that the parameter $z_{nl\,\mathrm{norm}}$ for the $5d$ and $4f$ elements is proportional to the degree of freedom of electrons (or holes) in the ground-state configuration.

ACKNOWLEDGMENTS

We would like to thank Dr. Y. Yano at RIKEN for his constant interest and support during this study and Dr. K. Morita for his useful discussions and comments. One of us (W.G.J.) would like to acknowledge the special researchers' basic science program.

[1] E. W. Otten, in *Treatise on Heavy-Ion Science,* edited by D. A. Bromley (Plenum, New York, 1989), Vol. 8, p. 517.
[2] D. L. Clark, M. E. Cage, D. A. Lewis, and G. W. Greenless, Phys. Rev. A **20**, 239 (1979).
[3] V. Pfeufer, W. J. Childs, and L. S. Goodman, J. Opt. Soc. Am. B **1**, 34 (1984).
[4] W. J. Childs, Phys. Rev. A **39**, 4956 (1989).
[5] J. Persson, U. Berzinsh, T. Nilsson, and M. Gustavsson, Z. Phys. D **23**, 67 (1992).
[6] A. Anastassov, Yu. P. Gangrsky, B. K. Kul'djanov, K. Marinova, B. N. Markov, E. Nadjakov, and S. G. Zemlyanoi, Z. Phys. D **25**, 1 (1992).
[7] J. Bauche and R.-J. Champeau, Adv. At. Mol. Phys. **12**, 39 (1976).
[8] H.-D. Kronfeldt, D. Ashkenasi, and H. M. Nikseresht, Z. Phys. D **22**, 569 (1992).
[9] G. Sawatzky and R. Winkler, Z. Phys. D **14**, 9 (1989).
[10] T. Brenner, S. Büttgenbach, W. Rupprecht, and F. Träber, Nucl. Phys. **A440**, 407 (1985).
[11] A. Nunnemann, D. Zimmermann, and P. Zimmermann, Z. Phys. A **290**, 123 (1979).
[12] D. Zimmermann, P. Zimmermann, G. Aepfelbach, and A. Kuhnert, Z. Phys. A **295**, 307 (1980).
[13] A. Kuhnert, A. Nunnemann, and D. Zimmermann, J. Phys. B **16**, 4299 (1983).
[14] Charles M. Miller, Rolf Engleman, Jr., and Richard A. Keller, J. Opt. Soc. Am. B **2**, 1503 (1985).
[15] Bryan L. Fearey, Denise C. Parent, Richard A. Keller, and Charles M. Miller, J. Opt. Soc. Am B **7**, 3 (1990).
[16] S. Büttgenbach, R. Dicke, H. Gebauer, R. Kuhnen, and F. Träber, Z. Phys. A **283**, 303 (1977).
[17] S. Büttgenbach, R. Dicke, and F. Träber, Phys. Rev. A **19**, 1383 (1979).
[18] R.-J. Champeau and M. Miladi, J. Phys. (Paris) **35**, 105 (1974).
[19] G. Gluck, Ann. Phys. (Paris) **10**, 673 (1965).
[20] N. K. Odintsova and A. R. Striganov, Opt. Spektrosk. **43**, 611 (1977) [Opt. Spectrosc. (USSR) **43**, 365 (1977)].
[21] P. Aufmuth, A. Steudel, and E. Wöbker, J. Phys. B **21**, 2237 (1988).
[22] P. Aufmuth, A. Steudel, W. Tegtmeier, and E. Wöbker, J. Phys. B **21**, 2253 (1988).
[23] M. Wakasugi, W. G. Jin, T. T. Inamura, T. Murayama, T. Wakui, T. Kashiwabara, H. Katsuragawa, T. Ariga, T. Ishizuka, M. Koizumi, and I. Sugai, Rev. Sci. Instrum. **64**, 3487 (1993).
[24] *Atomic Energy Levels, the Rare Earth Elements,* edited by W. C. Martin, R. Zalubas, and L. Hagan, Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 60 (U.S. GPO, Washington, DC, 1978).
[25] *Atomic Energy Levels,* edited by C. E. Moore, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, DC, 1958), Vol. III.
[26] H. Kopfermann, *Nuclear Moments, Pure and Applied*

Physics 2 (Academic, New York, 1958).
[27] K. Heilig and A. Steudel, At. Data Nucl. Data Tables **14**, 613 (1974).
[28] E. C. Seltzer, Phys. Rev. **188**, 1916 (1969).
[29] W. H. King, *Isotope Shifts in Atomic Spectra* (Plenum, New York, 1984).
[30] S. Büttgenbach, *Hyperfine Structure in 4d- and 5d-Shell Atoms* (Springer, Berlin, 1982).
[31] S. A. Ahmad, W. Klempt, R. Neugart, E. W. Otten, P.-G. Reinhard, G. Ulm, and K. Wendt, Nucl. Phys. **A483**, 244 (1988).
[32] S. A. Blundell, P. E. G. Baird, C. W. P. Palmer, D. N. Stacey, G. K. Woodgate, and D. Zimmermann, Z. Phys. A **321**, 31 (1985).
[33] K. Heilig, Hyp. Int. **24-26**, 349 (1985).
[34] P. Aufmuth, K. Heilig, and A. Steudel, At. Data Nucl. Data Tables **37**, 455 (1987).
[35] W. G. Jin, H. Sakata, M. Wakasugi, T. Horiguchi, and Y. Yoshizawa, Phys. Rev. A **42**, 1416 (1990).
[36] P. Aufmuth, R. Kirsch, A. Steudel, and E. Wöbker, Z. Phys. D **7**, 153 (1987).
[37] P. Aufmuth and E. Wöbker, Z. Phys. D **2**, 131 (1986).
[38] P. Aufmuth and E. Wöbker, Z. Phys. A **321**, 65 (1985).
[39] W. Neu, G. Passler, G. Sawatzky, R. Winkler, and H.-J. Kluge, Z. Phys. D **7**, 193 (1987).
[40] P. Aufmuth, A. Bernard, E.-G. Kopp, and A. Steudel, Z. Phys. D **27**, 1 (1993).