Optical isotope shifts of stable hafnium atoms in a resonance cell on-line with a heavy-ion accelerator

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Optical isotope shifts were measured on the 618-nm, ground-state transition of Hf I for the isotopes $^{174-180}$Hf. For the isotope pair 178-180, a specific mass shift of approximately -8 times the normal mass shift was deduced for this transition. The large size of this shift is consistent with other specific mass shifts that result from a broken d-shell electron pair. A transtion electronic factor of $E_{618}=0.158\pm0.012$ was extracted from the data. Excited-state hyperfine coefficients for the odd isotopes ^{179}Hf and ^{177}Hf were also deduced. The values of mean-square-charge radii obtained for the isotopic chain agree well with previously published results.

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I. INTRODUCTION

Studies of optical hyperfine spectra and isotope shifts of radioactive atoms have yielded a wealth of information about nuclear moments and changes in mean-square-charge radii $\lambda^{A,A'}\approx\delta\langle r^2\rangle$ along isotopic chains extending far from the line of stability [1]. The principal tools of such studies have been the on-line isotope separators used in conjunction with sensitive collinear laser spectroscopy. However, for the study of nuclides produced at low rates or from refractory elements, an on-line gas cell can be more suitable. The slow diffusion of the atoms within the gas allows a long interaction time with the laser, and hence an improved signal rate. Such a cell has already been used to study Yb atoms formed from unstable nuclei produced at rates as low as 10^3 s^{-1} [2,3], and now it has been used for studies of the refractory element hafnium.

The measurements reported here are of the optical spectra of the stable isotopes of hafnium. This work is preliminary to more extensive studies of radioactive atoms of hafnium. Working with the stable atoms has allowed us to become familiar with the atomic system and the isotope-shift parameters, a necessary prerequisite for attempting measurement of the unstable isotopes.

II. EXPERIMENT

Isotopes of stable hafnium nuclei have been accelerated by the SUNY Stony Brook Tandem Van de Graaff and injected through a thin Ni foil into the center of a liquid-nitrogen cooled gas cell. The cell and the method of on-line laser spectroscopy of accelerator produced atoms are described in detail in Ref. [4].

In one experiment, approximately 20–50 pA of 8+ Hf isotopes were accelerated to 30 MeV and implanted into $\approx$30 Torr of argon, to which was added 60 mTorr of N_2 and 12 mTorr of H_2 gases. In another experiment, 50 pA of 9+ Hf isotopes were accelerated to 38 MeV, but were implanted into only 8 Torr of argon containing a mixture of 60 mTorr of N_2 and 30 mTorr of H_2 gases.

The Hf atoms in the cell were illuminated with 27.5 mW/cm^2 of linearly polarized laser light, collimated into an 8-mm-diam beam which passed through the center of the cell. The laser beam was produced by a stabilized Spectra Physics tunable dye laser. Of the many possible transitions in Hf I that start from the ground state [5], we studied the one at 618.5 nm. This transition to the $^3D_2$ state at 16 163-cm^{-1} has one of the most favorable branching ratios down to the 2357-cm^{-1} level. The decay proceeds via 724.1-nm radiation (Fig. 1).

Off-resonant detection of this decay allowed us to greatly reduce background from scattered laser light. The detector, a photomultiplier, was masked with a narrow-band interference filter centered at 724 nm, and

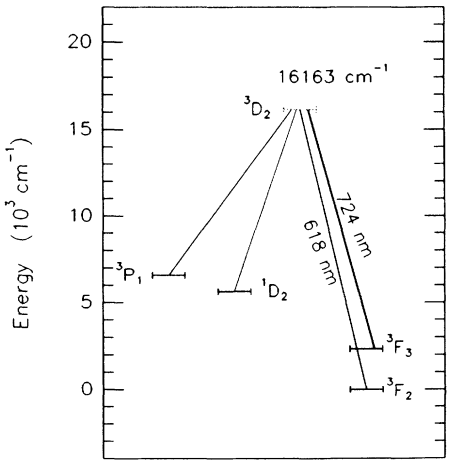

FIG. 1. Simplified level scheme of Hf I, showing the 618.5-nm transition from the ground state to the 16 163-cm^{-1} excited state and all known decays from this state including the strong 724.3-nm transition used in the present experiment to detect resonance.

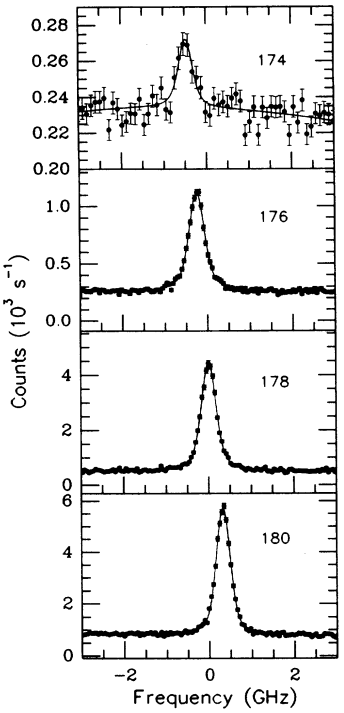

FIG. 2. Raw data scans of the 618-nm line for the even-mass hafnium isotopes. Fits to the data are indicated by the solid line.

this attenuated by about four orders of magnitude any 618-nm light reaching the detector. The incoming beam was pulsed, and by gating the signals on only when the ion beam was off it was possible to avoid counting background photons produced when ions from the accelerator entered the gas cell.

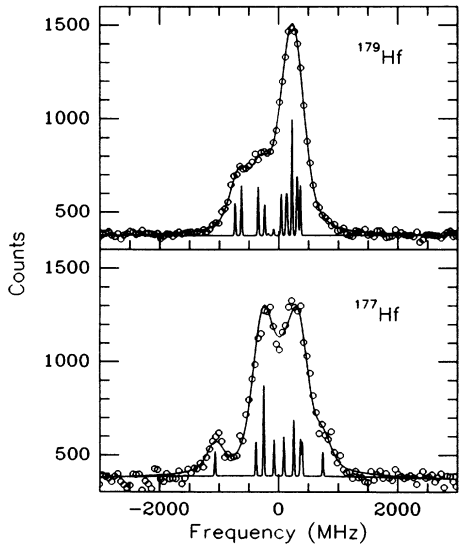

FIG. 3. Hyperfine structure of ^{177}Hf and ^{179}Hf. The data points show the measured line profile. The solid lines are fits to the data obtained as explained in the text. Also shown are the positions and relative intensities of the strongest of the 13 lines of the hyperfine multiplet.

A plot of photomultiplier counts versus laser frequency yields the line profiles shown in Figs. 2 and 3. Because the atoms stop in a background gas of several Torr, their linewidths are both Doppler and pressure broadened, with typical full width at half maximum on the order of 250 MHz. The line profiles were fit by a Voigt profile using a multiparameter least-squares-fitting routine, assuming a known Doppler width derived from the temperature of the gas. The line centroid of each isotope, corrected for a measured pressure shift of -3.97 ± 0.34 MHz/Torr, was then compared to a common frequency scale. This scale was established from an iodine spectrum that was taken concurrently with the Hf spectra and corrected for scanning nonlinearities with the aid of a marker spectrum produced with a high finesse, 2-GHz étalon.

The addition of N_2 and H_2 impurity gases to the argon buffer gas was crucial to the experiment. Adding them increased the fluorescent yield from each atom by about a factor of 200. This effect has been explained, both qualitatively and quantitatively, by a semiclassical model based on simple rate equations for all the energy levels involved in the transition and decay [6]. This model will be discussed in a later paper.

III. RESULTS

The centroids of the even-mass isotopes, shown in Fig. 2, were determined from fits as described above. However, the odd-mass isotopes, ^{179}Hf and ^{177}Hf ($I^{\pi}=\frac{9}{2}^{+}$ and $\frac{7}{2}^{+}$, respectively), display complex line shapes due to hyperfine splitting in both the ground and excited states. The measured line profiles are sums of 13 discrete hyperfine lines of various intensities, each broadened with a full Voigt profile as shown in Fig. 3.

The individual line contributions were determined from the complex profiles by using the *ground-state A* and *B* hyperfine coefficients to calculate the hyperfine line frequencies. These coefficients had been previously measured to high precision [7]. From assumed trial values of the excited-state coefficients, all transition frequencies were calculated. The intensity of each line was calculated for linearly polarized excitation, assuming no optical pumping of the hyperfine states.

Each trial frequency was then broadened with the Voigt profile extracted from the even-isotope data, and the intensity profiles of all lines were summed to reproduce the measured line shape. In this way the measured line was "fit" by a theoretical line profile, and in the process the excited-state *A* and *B* parameters, as well as the line centroid, were extracted.

The remarkably good fits are shown as the solid lines in Fig. 3 with an uncertainty in the determination of the *A* and *B* coefficients of roughly 1%. From the known values of the nuclear magnetic dipole and nuclear electric quadrupole moments [8], the mean magnetic field $B(0)$ and the electric-field gradient eV_{zz} at the nucleus were deduced for the 16 163-cm^{-1} level. The results are summarized in Table I. For the two isotopes the intensities of the lines and their positions relative to the line centroid are listed in Table II; they have uncertainties of a few percent. The good agreement between the two isotopes for the deduced values of $B(0)$ and eV_{zz}, despite greatly

TABLE I. Hyperfine coefficients and other nuclear parameters of the 16 163-cm^{-1} excited state of the odd-mass hafnium isotopes.

Atomic mass (amu)	Nuclear magnetic dipole moment μ_I (nm)	Magnetic hyperfine coefficient A (MHz)	Mean magnetic field $\overline{B(0)}$ (kG)	Nuclear electric quadrupole moment Q b	Quadrupole hyperfine coefficient B (MHz)	Mean electric field $\overline{eV_{zz}}$ (eV/cm^2)
177	0.7836(6)	75.6(6)	675(5)	3.30(65)	685(17)	207(41)
179	−0.6329(13)	−47.8(6)	680(9)	3.72(74)	756(17)	203(41)

different hyperfine and nuclear parameters, lends credence to the fits and the determination of the line centroids. If, however, optical pumping of the hyperfine states does occur, then these results and those that follow would have to be modified.

Isotope shifts were then calculated from the zero-pressure centroids of all lines. Figure 4 is a plot of these shifts relative to ^{178}Hf as a function of isotope mass. The plot shows a small positive shift of roughly $\delta\nu^{A,A+2}\approx 300$ MHz. Since no s electron is involved in the transition (the 618-nm resonance line is a transition out of the ground-state $5d^26s^2\ ^3F_2$ configuration, to an excited-state configuration of $5d6s^26p\ ^3D_2$ at 16 163 cm^{-1}), such small shifts are to be expected.

Table III lists the frequency shifts for all isotopes, as well as a breakdown of the various contributions. All shifts are with respect to ^{178}Hf. The normal mass shift $\delta\nu_{\rm NMS}$ can be calculated unambiguously as

$$\delta\nu_{\rm NMS}=\frac{\nu_i}{1836.1}\frac{A'-A}{A'A}\ , \tag{1}$$

with $\nu_i=c/\lambda=2.9979\times10^{11}/618.5$ MHz. The specific mass shifts $\delta\nu_{\rm SMS}$ were extracted using King plots [9], which require isotope shifts from two different transitions. Our measured isotope shifts were compared with those of Čajko [10], who reports on 25 transitions in which shifts were measured between three pairs of isotopes. Twenty of those transitions yielded statistically meaningful fits, and King plots were made with respect to these lines. Some of the plots are shown in Fig. 5. All of the lines are $6s^2$-$6s6p$ transitions, which typically have small specific mass shifts. As in Refs. [11] and [12], they were taken to have $\delta\nu_{\rm SMS}=(0.0\pm0.5)\delta\nu_{\rm NMS}$. Using the slope and the intercept from the King plot, we have extracted the $\delta\nu_{\rm SMS}$ for the 618-nm transition and obtained the values shown in Fig. 6. For the isotope pair 180-178, the shift is

$$\delta\nu_{\rm SMS}=-139\pm12\ \text{MHz}\ .$$

This is to be compared with the normal mass shift of $\delta\nu_{\rm NMS}=16.5$ MHz. The negative sign indicates that the correlated electron motion tends to overcompensate the

TABLE II. Lines, positions, and normalized intensities of the hyperfine multiplets of the 618.5-nm transitions in ^{177}Hf and ^{179}Hf. The total angular momentum of the ground and excited states are denoted by $F_{\rm gnd}$ and $F_{\rm ex}$, respectively. $\Delta\nu$ is the displacement of the component from the center of mass of the line; Int is the intensity relative to the strongest transition.

	^{177}Hf				^{179}Hf			
Line	$F_{\rm ex}$	$F_{\rm gnd}$	$\Delta\nu$ (MHz)	Int	$F_{\rm ex}$	$F_{\rm gnd}$	$\Delta\nu$ (MHz)	Int
1	$\frac{3}{2}$	$\frac{3}{2}$	373.2	0.170	$\frac{5}{2}$	$\frac{5}{2}$	−273.1	0.265
2	$\frac{3}{2}$	$\frac{5}{2}$	368.4	0.254	$\frac{5}{2}$	$\frac{7}{2}$	321.5	0.291
3	$\frac{5}{2}$	$\frac{3}{2}$	256.4	0.254	$\frac{7}{2}$	$\frac{5}{2}$	−735.2	0.291
4	$\frac{5}{2}$	$\frac{5}{2}$	251.6	0.003	$\frac{7}{2}$	$\frac{7}{2}$	−176.5	0.018
5	$\frac{5}{2}$	$\frac{7}{2}$	88.7	0.378	$\frac{7}{2}$	$\frac{9}{2}$	365.4	0.433
6	$\frac{7}{2}$	$\frac{5}{2}$	259.2	0.378	$\frac{9}{2}$	$\frac{7}{2}$	−627.8	0.433
7	$\frac{7}{2}$	$\frac{7}{2}$	96.3	0.081	$\frac{9}{2}$	$\frac{9}{2}$	−85.9	0.056
8	$\frac{7}{2}$	$\frac{9}{2}$	−380.7	0.388	$\frac{9}{2}$	$\frac{11}{2}$	307.0	0.439
9	$\frac{9}{2}$	$\frac{7}{2}$	399.7	0.388	$\frac{11}{2}$	$\frac{9}{2}$	−348.5	0.439
10	$\frac{9}{2}$	$\frac{9}{2}$	−77.3	0.401	$\frac{11}{2}$	$\frac{11}{2}$	44.4	0.375
11	$\frac{9}{2}$	$\frac{11}{2}$	−1069.1	0.269	$\frac{11}{2}$	$\frac{13}{2}$	126.5	0.300
12	$\frac{11}{2}$	$\frac{9}{2}$	741.8	0.269	$\frac{13}{2}$	$\frac{11}{2}$	143.7	0.300
13	$\frac{11}{2}$	$\frac{11}{2}$	−250.0	1.000	$\frac{13}{2}$	$\frac{13}{2}$	225.8	1.000

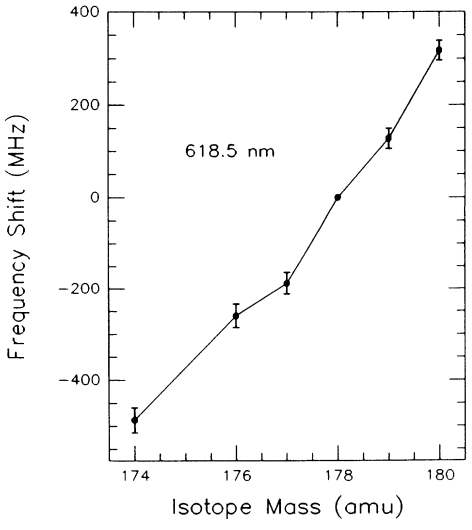

FIG. 4. Isotope shifts of the 618-nm line relative to ^{178}Hf.

TABLE III. Isotope shifts and extracted $\lambda^{A,A'}$ for the 618-nm line of the stable hafnium isotopes. The reference isotope is ^{178}Hf.

A	A'	$\delta\nu^{A,178}$ (MHz)	$\delta\nu_{NMS}$ (MHz)	$\delta\nu_{SMS}$ (MHz)	$\delta\nu_{MS}$ (MHz)	$\delta\nu_{FS}$ (MHz)	λ (fm^2)
174	178	487(27)	34.2	−288(24)	−254(24)	741(36)	0.117(10)
176	178	259(26)	16.9	−142(12)	−126(12)	385(29)	0.061(6)
177	178	188(24)	8.4	−71(6)	−62(6)	250(25)	0.040(5)
178	179	128(22)	8.3	−70(6)	−62(6)	190(23)	0.030(4)
178	180	317(21)	16.5	−139(12)	−123(12)	440(24)	0.070(6)

effect of the valence electron for this transition. The total mass shifts are listed in column three of Table III. Subtracting these values from the total shift $\delta\nu^{A,178}$ yields the field shifts in column four.

Since the relative shifts and $\lambda^{AA'}$ for the hafnium isotopes have been previously measured to high precision [10,13–15], we were able to deduce the transition electronic factor E_{618} (assuming no mass or second-order J dependence of the field shift). The transition electronic factor found from the field shift is

$$E_{618}=\frac{\delta\nu_{FS}^{A,A'}}{f(Z)\lambda^{A,A'}},$$

where $f(Z)$ is a known function [16]. The values of $\lambda^{A,A'}$, relative to ^{178}Hf are, respectively [11],

174	176	177	179	180
−0.120(17)	−0.060(9)	−0.043(6)	0.025(4)	0.069(10)

The units are fm^2. The mean value for the electronic factor was determined to be

$$E_{618}=0.158\pm0.012\ .$$

The consistency of the $\delta\nu_{SMS}$ extraction was then checked by calculating $\lambda^{A,178}$ using our isotope shifts and the above value for E_{618}. The results are listed in the last column of Table III. The large value of the specific mass shift significantly alters the mass shift's contribution to the isotope shift, and thus affects the value of the field shift that is extracted. A naive assumption for the specific mass shift of $\delta\nu_{SMS}=0.0$ would have resulted in an underestimation of E_{618} by about 30%, and a subsequent underestimation of $\lambda^{A,A'}$ derived from future experiments.

IV. CONCLUSION

Initial investigations of the hafnium isotopes using an on-line gas cell have resulted in the determination of isotope-shift parameters of the 618-nm transition in Hf I. These include the electronic transition factor E_{618}, and the specific mass shift. The latter is interesting because of its relatively large value and because of its overall effect on the determination of $\lambda^{A,A'}$. The size of the contribution is not expected for heavy isotopes, although for transitions in which a d-shell electron pair is broken, the large value is not without precedent [12,17]. Values were also obtained for the excited-state A and B hyperfine coefficients and for the pressure shift of the 618-nm transition in argon.

It appears that the prospects for measuring isotope

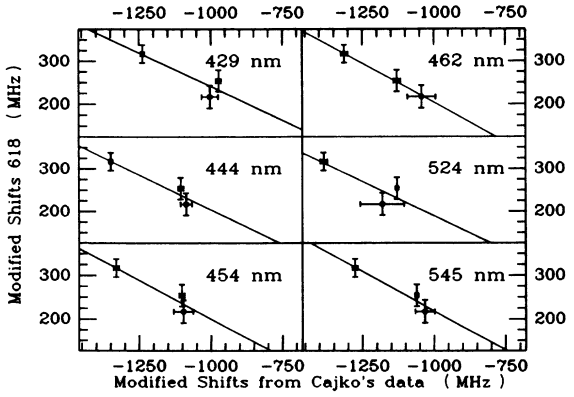

FIG. 5. King plots of the 618-nm line vs lines taken from Ref. [10]. The plotted shifts are modified as described in Ref. [9], p. 63. For display purposes, all values have been multiplied by 2/(180) (178).

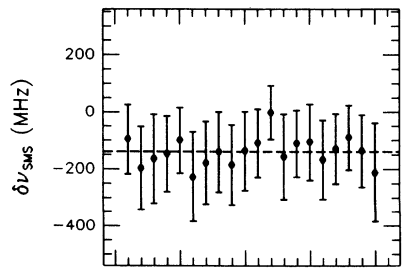

FIG. 6. The values of the SMS obtained from the above King plots. On the x axis is plotted an index number that counts the transitions. The dashed line corresponds to the mean value of the shifts, −139±12 MHz.

shifts of atoms of unstable nuclei of hafnium using the on-line gas cell are good, as are the possibilities of measuring other parameters relevant to the atomic and nuclear physics of hafnium.

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