Nuclear Properties of the Exotic High-Spin Isomer $^{178}Hf^{m2}$ from Collinear Laser Spectroscopy

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The complete hyperfine spectrum in the optical transition $5d^26s^2\,{}^3P_2 \rightarrow 5d6s^26p\,{}^1P_1$ of $^{178}Hf^{m2}$ was recorded by collinear laser spectroscopy using nanogram amounts of samples. The quadrupole moment and isomer shift were determined for the first time as well as a precise value and the sign of the magnetic dipole moment. The change in nuclear mean-square charge radius between the isomeric state $^{178}Hf^{m2}$ and the ground state $^{178}Hf^{g}$ was evaluated as $\delta\langle r^2\rangle^{178,178m2} = -0.059(9)$ fm^2. From the hyperfine A and B factors the magnetic moment $\mu I^{78m2} = +8.16(4)$ nuclear magnetons and the spectroscopic quadrupole moment $Q_s^{178m2} = +6.00(7)$ b were extracted.

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When an excited state in an atomic nucleus can decay to lower lying states only by γ ray transitions these transitions may be considerably hindered, if the difference in angular momentum between the excited state and all lower lying states is large. Thus, the ground state angular momentum being $I=0$ in the nuclei with an even number of protons and neutrons, isomeric states with large angular momenta can appear at relatively low excitation energy, often considerably lower than the states of similar I values of the ground state rotational (or yrast) band. A large number of such high-spin isomers are observed in the region of the rare earth elements and beyond. Their high-spin values are interpreted as due to the alignment of the angular momenta of several nucleons. In this respect, the nucleus ^{178}Hf is very interesting since it shows the four-quasiparticle configuration $^{178}Hf^{m2}$ with spin $I^\pi = 16^+$ [1] and the extremely long half-life $T_{1/2} = 31$ yr [2]. Since this state has the lowest excitation energy $E = 2447.4$ keV for its angular momentum I and is located even below the $I=14$ levels, it cannot decay by low multipolarity transitions. The exceptionally long half-life even allows the preparation of nanogram amount of samples and targets of this excited nuclear matter for the present optical measurements as well as for inelastic scattering and Coulomb excitation experiments [3]. Thus, the nuclear properties can be studied in two states: the isomer with its four unpaired nucleons and the ground state with its fully paired structure ($I=0$).

In the decay of the $I^\pi = 16^+$ state a $T_{1/2} = 4s$ isomer with $I^\pi = 8^-$ at 1147.4 keV is populated and a second $I^\pi = 8^-$ level is located at 1479.0 keV. These two states are interpreted as mixtures of the two proton configuration $\{p[514]9/2 + p[404]7/2\} I^\pi = 8^-$ ($K=8$, K being the projection of **I** on the nuclear symmetry axis) and the two neutron configuration $\{n[514]7/2 + n[624]9/2\} I^\pi = 8^-$ ($K=8$) [2,4]. The sum of excitation energies of these two states is close to the energy of the long-lived isomer. This suggests that the $I^\pi = 16^+$ ($K=16$) state is a superposition of the two 8^- configurations and thus of a pure four-quasiparticle nature.

In this Letter, we present optical measurements on $^{178}Hf^{m2}$ together with corresponding systematic results obtained for the stable isotopes $^{176\text{-}180}Hf$. The measurements provide the A and B factors for the atomic levels of the transition which allow determination of the magnetic moment μ_I and the spectroscopic quadrupole moment Q_s [5]. The isomer shift (IS) between the center of gravity of the hyperfine spectrum (HFS) of the isomeric state and the ground state resonance line allows determination of the change in the mean-square nuclear charge radius $\delta\langle r^2\rangle^{178,178m2}$ between the isomer and the ground state. These results include the first measurement of the quadrupole moment and the isomer shift as well as a more precise value and the sign of the magnetic dipole moment of $^{178m2}Hf$. The measurements are particularly challenging due to the extremely low production rate of the exotic Hf isomer that results in a microscopic sample.

The Hf isomer was produced in the reaction $^{176}Yb(\alpha, 2n)$ by bombarding a 96% enriched Yb target with a 36 MeV α beam provided by the U-200 cyclotron at Dubna [6]. The two samples for our experiments were chemical-

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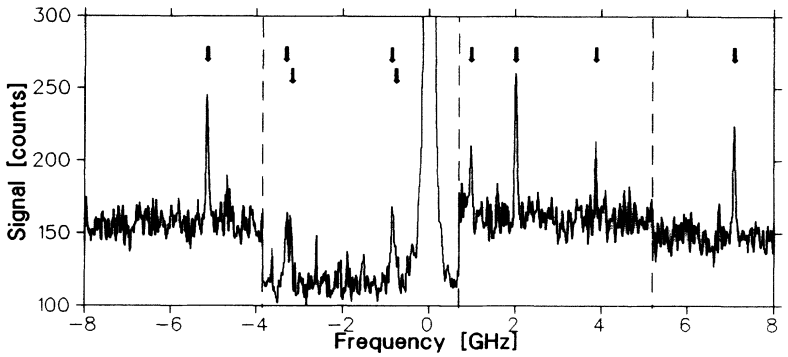

FIG. 1. Experimental hyperfine spectrum of ^{178}Hf. The spectrum was recorded over four different ranges as shown. The single peak at 0 MHz having an amplitude of 14000 counts corresponds to the ground state ^{178}Hf^{g}. The nine hyperfine resonances of ^{178}Hf^{m2} are marked by arrows.

ly separated and deposited on a quartz substrate. Each of the samples contained about 2×10^{13} atoms or 6 ng of ^{178m2}Hf and a roughly 30 times larger amount of ^{178g}Hf as deduced from the optical signals. By chlorination the sample was volatilized and fed into the ion source of the P.A.R.I.S. mass separator [7] at the CSNSM at Orsay. The 40 keV elemental ion beam was mass separated and transported to a collinear laser spectroscopy apparatus where the ions were neutralized in a charge exchange cell with sodium vapor and merged with a counterpropagating laser beam of typically 15 mW from a single-mode dye laser. This dye laser was stabilized to a molecular absorption line of I_2. Spectroscopy was performed by exciting the $5d^26s^2\,{}^3P_2$ (8983.75 cm^{-1}) $\rightarrow$ $5d6s^26p\,{}^1P_1$ (26463.93 cm^{-1}) transition ($\lambda=572$ nm) and then detecting the fluorescence of the subsequent decay through the strong resonance line ($\lambda=378$ nm) to the $5d^26s^2\,{}^3F_2$ ground state. The hyperfine resonances were scanned via the beam velocity by applying a sweep voltage to the charge exchange cell. An axial magnetic field of 2×10^{-2} T was applied to the charge exchange cell and a subsequent drift space in order to suppress optical pumping outside the detection zone. A large solid angle ellipsoidal mirror system has been used to collect the fluorescence light to a photon counting RCA8850 photomultiplier. An interference filter and a Schott BG3 color glass were inserted between the collection system and the entrance window of the photomultiplier. Details are given in [8] but special care has been taken in order to reduce beam background signal due to scattering of the strong ^{178g}Hf beam to the detection system. A signal/background ratio of 400 has been achieved for stable even isotopes.

The measured hyperfine spectrum of ^{178}Hf^{m2+g} is shown in Fig. 1. The single peak corresponds to the $I^\pi=0^+$ ground state. The positions were fitted to the HFS formula [9] yielding the hyperfine A and B factors for the lower $^3P_2$ and upper $^1P_1$ level as well as the isomer shift in the optical transition. In addition we have recorded hyperfine spectra of this transition for the stable Hf isotopes in order to determine their A and B factors and isotope shift needed for the extraction of the atomic parameters. The results are listed in Table I.

The isotope shift (S_I) can be decomposed into the normal mass shift (S_{NM}), the specific mass shift (S_{SM}), and the field shift (S_F). S_F is expressed by $S_F^{A,A'}=F_i\lambda^{A,A'}$ where F_i is the electronic factor for the transition and the nuclear parameter $\lambda^{A,A'}=K\delta\langle r^2\rangle^{A,A'}$, where $\delta\langle r^2\rangle^{A,A'}$ stands for the change in the mean-square charge radius of the two nuclei under consideration (for Hf, $K=0.95$ [10]). The nuclear parameter $\lambda^{178,180}$ has been obtained by a parametric analysis of 33 transitions in Hf II [11] to be $\lambda^{178,180}=+0.072(4)$ fm^2 which, together with the model independent relative nuclear parameters λ_{rel} [12], produces the values in Table I for the stable isotopes. S_{SM}(572 nm) and $F_{572\,nm}$ have been determined by a

TABLE I. Hyperfine constants of the $5d^26s^2\,{}^3P_2$ and $5d6s^26p\,{}^1P_1$ atomic states, the isotope shift $S_I^{178,A}$, and the nuclear parameter $\lambda^{178,A}$ for ^{178}Hf^{m2} and the stable isotopes $^{176-180}$Hf.

A (amu)	I^π	$A(^3P_2)$ (MHz)	$A(^1P_1)$ (MHz)	$B(^3P_2)$ (MHz)	$B(^1P_1)$ (MHz)	$S_I^{178,A}$ (MHz)	$\lambda^{178,A}$ (fm^2)
176	0^+	...	...	...	...	−121.9(5.3)	−0.063(4)
177	$7/2^-$	+70.9(4)	−21.9(6)	−1206.5(3.7)	+687.7(2.0)	−87.1(3.0)	−0.045(3)
178	0^+	...	...	...	...	0.00	0.000
178 m2	16^+	+159.5(2)	−51.3(4)	−2150.7(16.2)	+1234.8(9.6)	−134.6(3.1)	−0.056(8)
179	$9/2^+$	−44.7(2)	+13.9(2)	−1364.0(2.4)	+776.3(1.5)	...	+0.027(2)
180	0^+	...	...	...	...	+133.9(5.1)	+0.072(4)

King plot [13], which relates the S_I for one transition to that of another; in this case the 618 nm transition of Hf I [14]. $S_{SM}(618\text{ nm})$ and $F_{618\,nm}$ were, in turn, evaluated by King plot procedures using 13 transitions between the ground state multiplet $5d^26s^2\,{}^3F$ and $5d^26s6p$ configurations investigated by Cajko [15]. Since these configurations are pure [16], the S_{SM} in these transitions can be estimated to be nearly zero [12]. Their electronic factors have been obtained by a fit of the measured $S_I^{A,A'}$ to the $\lambda^{A,A'}$ from Table I. Finally, we determined $F_{572\,nm}=+2396(347)$ MHz/fm^2 and $S_{SM}(572\text{ nm})=-3.00(1.09)\,S_{NM}(572\text{ nm})$. (There are muonic data available [17] for the calibration of the electronic factor. $F_{572\,nm}$ calibrated in this way differs by about -35% from the above value.) For ^{178}Hf, the change in the mean-square charge radius between the fundamental and the isomeric state is thus determined to be

$$\delta\langle r^2\rangle^{178,178m2}=-0.059(9)\text{ fm}^2 \quad (1)$$

which almost corresponds to the subtraction of a neutron pair. The isotonic ^{177,177}Lu^m also exhibits a negative isomer shift but of a lower absolute value [18].

The absolute value of $\delta\langle r^2\rangle^{178,178m2}$ and its negative sign are very difficult to interpret. Because of the different pairing properties for the 0^+ and 16^+ states, there are a variety of contributions to this quantity. These contributions include monopole, quadrupole, hexadecapole, and higher moments as well as coupling between the various modes. Moreover, current theoretical approaches generally do not deal with wave functions projected on good angular momentum values which immediately make them unsuitable for the description of small variations.

From the magnetic moments of ^{177,179}Hf [19] and the $A(^3P_2)$ factors in Table I the hyperfine anomaly [20] is found to be $^{177}\Delta^{179}(^3P_2)=-3.6(7.5)\times10^{-3}$, which is comparable to the corresponding value for the ground state $^{177}\Delta^{179}(^3F_2)=-2.4(2.2)\times10^{-3}$ [21]. Thus, neglecting hyperfine anomaly with respect to experimental uncertainties, the magnetic moment of ^{178}Hf^{m2} can be scaled according to the relation $A=\mu_I\langle H(0)\rangle/(IJ)$. With the accurate value for μ_I^{177} [19] and the A factors from Table I we obtain

$$\mu_I^{178m2}=+8.16(4)\mu_N\,, \quad (2)$$

where μ_N denotes the nuclear magneton. An earlier rough measurement via nuclear orientation [22] yielded $|\mu_I^{178m2}|=7.26(+20)(-00)\mu_N$.

The magnetic moment of a nuclear state is mainly due to its single-particle properties. The modified oscillator model [23] coupling the g_{K_i} values of the nuclear states involved has given a theoretical value of $\mu_{theo}^{178m2}=+7.81\mu_N$. Combining the experimental moments of neighboring odd nuclei by the coupling rules given in [23] yields a semiempirical value $\mu_{SE}^{178m2}=+8.17\mu_N$ in excellent agreement with the experimental result (2), which further confirms the four-quasiparticle assignment for the isomeric state [2].

For ^{177}Hf, a spectroscopic quadrupole moment $Q_s=3.365(29)$ b has been obtained from muonic data [19], which is comparable to $Q_s=3.30(65)$ b (including Sternheimer correction) from *ABMR* measurements [20]. To determine Q_s for the isomer we use the first value for ^{177}Hf and scale with the B $(5d^26s^2\,{}^3P_2)$ factors (Table I) to obtain

$$Q_s^{178m2}=+6.00(7)\text{ b}\,. \quad (3)$$

Assuming the strong coupling scheme for evaluating the intrinsic quadrupole moment from the measured Q_s (3), we obtain a value of $Q_0=7.2(1)$ b for the $I^\pi=16^+$ state. This is slightly higher than $Q_0=6.961(43)$ b for the ground state derived from $B(E2)$ values [24]. Theoretical calculations [25,26] have predicted $Q_0=7.3$ b for the ground state with very little variation for the 16^+ isomeric state.

Our collinear laser spectroscopy experiment has resulted in the determination of the magnetic dipole and electric quadrupole moment as well as the isomer shift of the high-spin isomer ^{178}Hf^{m2}. While the magnetic moment can be determined mainly by the single-particle nature of the nuclear states under consideration, the explanation for the negative isomer shift (and thus the change in mean-square charge radius) needs detailed microscopic calculations incorporating higher orders of deformation.

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