

Shrinking of the Nuclear Charge Radius in the First Rotational State of Hf¹⁷⁸

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The Mössbauer isomer shift for the 93.2-keV transition in Hf¹⁷⁸ has been studied in Hf metal, HfO₂, HfC, and cyclopentadienyl hafnium dichloride, Hf(Cp)₂Cl₂. One can qualitatively understand the shifts in terms of shielding of *s* electrons by valence *5d* electrons. An unambiguous isomer shift of $+0.19 \pm 0.06$ mm/sec has been observed between Hf(Cp)₂Cl₂ and Hf metal. This is interpreted as evidence of a shrinking of the nuclear charge radius of Hf¹⁷⁸ in the first rotational state. We estimate the change in the *s* electron density between Hf(Cp)₂Cl₂ and Hf metal by correlating it to the observed isomer shifts between Os(Cp)₂ and Os metal and calculate

$$\delta\langle r^2 \rangle(\text{Hf}^{178}) = -0.37 \times 10^{-3} \text{ fm}^2, \quad \delta\langle r^2 \rangle / \langle r^2 \rangle(\text{Hf}^{178}) = -0.13 \times 10^{-4}.$$

INTRODUCTION

In this paper we report on the measurement of the isomer shift of the 93.2-keV ($2^+ \rightarrow 0^+$) transition in Hf¹⁷⁸ using the Mössbauer effect. We have observed an unambiguous isomer shift between cyclopentadienyl hafnium dichloride Hf(Cp)₂Cl₂ and hafnium metal. From these measurements, the sign of the change in the mean square radius $\delta\langle r^2 \rangle$ has been found to be negative, thus indicating that the charge radius of Hf¹⁷⁸ in its first rotational state is smaller than in the ground state.

This result disagrees with a previous Mössbauer measurement,¹ where a positive sign of $\delta\langle r^2 \rangle$ was inferred from isomer-shift measurements between HfO₂ and Hf metal. We also performed experiments with HfO₂ and Hf metal but have found no measurable isomer shift between these absorbers. The smallness of the shift observed between HfO₂ and Hf metal on the one hand and the large shift observed for the Hf organometallic on the other, is consistent with isomer shift observations in other *5d* elements^{2, 3} like W, Os, and Ir. In all these cases, the isomer shift can be qualitatively understood in terms of the shielding of *s* electrons by valence *5d* electrons. The use of an organometallic compound in the present isomer shift study of even-even Mössbauer isotopes is novel and offers some distinct advantages.

The shrinking of the nuclear charge radius in the first rotational state of an even-even nucleus is a strange result. In the past, there have been Mössbauer measurements^{3, 4} and muonic x-ray studies⁵ that have suggested the same results.

The Mössbauer results are summarized and reviewed.

EXPERIMENTAL METHODS AND RESULTS

Mössbauer spectra were recorded in a conventional transmission geometry using a constant acceleration Mössbauer drive with a 512-channel analyzer, operated in the multiscaling mode. In all experiments, both sources and absorbers were cooled to liquid-helium temperature in an exchange-gas Dewar. The drive was calibrated using a Co⁵⁷ (Cu) source with enriched metallic iron and sodium nitroprusside absorbers.

20-day W¹⁷⁸, in tantalum metal, was used as a source of the 93.2-keV γ ray. Details of source production and heat treatment of the source foil to obtain a narrow emission linewidth are discussed in a separate paper.⁶ A study of line broadening of the nuclear γ resonance with cubic HfC absorbers of different effective thicknesses showed (a) the linewidth extrapolated to zero absorber thickness to be 1.90 ± 0.07 mm/sec which may be compared with the natural linewidth of 1.99 ± 0.04 mm/sec, deduced from the electronically measured lifetime,⁷ and (b) the Debye temperature of HfC to be (310 ± 14) °K. The experimental data are summarized in Fig. 1. The Mössbauer spectra taken with HfC and Hf(Cp)₂Cl₂ show a single line and those of Hf metal and HfO₂ show a partially resolved doublet.

HfC

The Mössbauer spectra taken with HfC absorbers were least-squares fitted to a single Lorent-

zian line shape. This yielded an isomer shift of -0.05 ± 0.03 mm/sec with respect to the tantalum metal source. An x-ray examination of our sample confirmed the fcc B1 structure and yielded the lattice parameter $a = 4.6349 \pm 0.0029$ Å in accord with the values reported in the literature.⁸

Hf Metal

The electric quadrupole interaction in Hf metal has been studied extensively by Mössbauer effect⁹ and perturbed angular-correlation technique.¹⁰ In our earlier Mössbauer experiments¹¹ on Hf metal, both in polycrystalline and single-crystal form, we showed that the electric quadrupole interaction in the hcp lattice has axial symmetry $\eta \sim 0$. For the purpose of extracting the isomer shift in the present studies, Mössbauer spectra of Hf metal were taken both in polycrystalline and single-crystal form. Figure 1 reproduces the spectrum taken with 99.9% pure Hf metal in a polycrystalline form. The spectrum was least-squares fitted to an axially symmetric electric field gradient (EFG) ($\eta = 0$), constraining the intensities of the quadrupole components to be in the ratio of 1:2:2. This fitting procedure gave the strength of the quadrupole interaction e^2qQ

TABLE I. Summary of Mössbauer results on the electric quadrupole interaction parameters using Hf¹⁷⁸. All measurements were made at liquid-helium temperature.

Lattice	e^2qQ (μeV)	η	Reference
Hf metal polycrystalline	-1.752 ± 0.024	0 ^a	9
Hf metal polycrystalline	-1.84 ± 0.01	0 ^a	1
Hf metal single and polycrystalline	-1.83 ± 0.06	~ 0 ^b	11
Hf metal single and polycrystalline	-1.85 ± 0.03	0 ^a	Present
HfO ₂ polycrystalline	-2.40 ± 0.02	0 ^a	9
HfO ₂ polycrystalline	-2.486 ± 0.007	0.475	1
HfO ₂ polycrystalline	-2.48 ± 0.03	0.31 ± 0.12	Present
HfO ₂ polycrystalline	-2.51 ± 0.03	0 ^a	
Hf(Cp) ₂ Cl ₂ polycrystalline	-1.10 ± 0.10	0 ^a	Present

^a Analysis made by constraining $\eta = 0$.

^b η was shown to be small ~ 0 .

$= 1.85 \pm 0.03$ μeV and an isomer shift of $+0.02 \pm 0.04$ mm/sec (Table II) with respect to the source. The line positions of the $|\Delta m| = 1$ and $|\Delta m| = 2$ quadrupole components obtained from the polycrystalline fit, coincided within experimental errors with those observed using Hf single-crystal absorber cut along the $[10\bar{1}0]$ axis. The quadrupole interaction strength measured for Hf metal agrees well with previous Mössbauer measurements (Table I).

HfO₂

The Mössbauer spectrum of HfO₂ was interpreted in terms of a partially resolved electric quadrupole interaction.^{1,9} Extraction of the isomer shift was carried out by least-squares fitting the spectrum in two ways, assuming the EFG to possess (a) axial symmetry ($\eta = 0$), and (b) non-axial symmetry ($\eta \neq 0$). A slightly better fit of the spectrum was possible under assumption (b), which yielded a value of $\eta = 0.31 \pm 0.12$ and $e^2qQ = (2.48 \pm 0.03)$ μeV . These measurements are compared with previous measurements in Table I, and the agreement is good. Both analyses yielded the same isomer shift within limits of experimental error and the shift was found to be $+(0.03 \pm 0.04)$ mm/sec (Table II). The crystal structure of HfO₂ is reported¹² to be monoclinic and the lattice parameters are $a = 5.1156(5)$ Å, $b = 5.1722(5)$

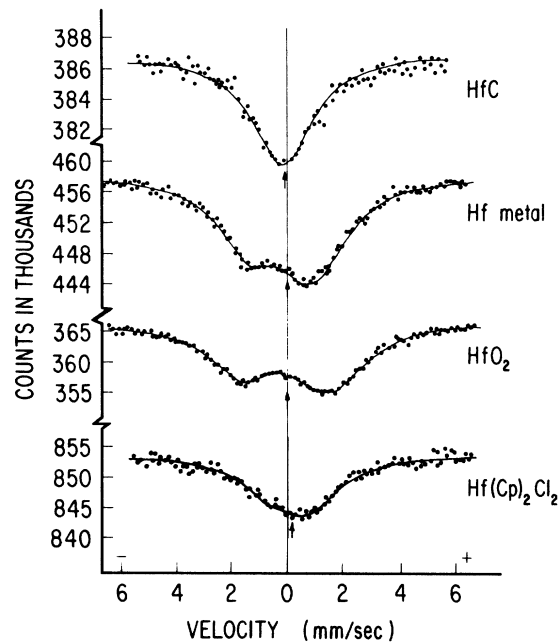


FIG. 1. Mössbauer spectra taken with W¹⁷⁸ source in tantalum metal using absorbers of: (a) HfC; (b) Hf metal; (c) HfO₂; and (d) Hf(Cp)₂Cl₂.

\AA , $c = 5.2948(5) \text{\AA}$, and $\beta = 99^\circ 11'$, $0^\circ 05'$. These data suggest that the existence of a finite asymmetry parameter would not be inconsistent with the known crystal structure.

Hf(Cp)₂Cl₂

Mössbauer spectra of Hf(Cp)₂Cl₂ absorbers¹³ were recorded for different thicknesses of the absorbers. In each case, the spectrum was least-squares fitted to a single Lorentzian line shape. This yielded an isomer shift of 0.21 ± 0.05 mm/sec with respect to the source. Since the stability of the absorber was unknown, all absorbers were prepared in an inert gas atmosphere (nitrogen) and measurements were carried out on two separate batches of samples. One of these batches was more than 2 yr old. The isomer shifts obtained using different samples were found to be reproducible. The spectra were fitted to both (a) a single line, and (b) an electric quadrupole interaction with $\eta = 0$ and $\eta \neq 0$. A better fit of the data is possible under assumption (b), shown in Fig. 1. We obtained $e^2qQ = -1.1 \pm 0.10$ eV assuming $\eta = 0$ (Table I). This is to be expected, since the structure of Hf(Cp)₂Cl₂ is not cubic. The wedge-shaped sandwich structure of Hf(Cp)₂Cl₂¹⁴ is similar to Ti(Cp)₂Cl₂ and Zr(Cp)₂Cl₂. The metal ion is bonded to the two cyclopentadienyl rings and chlorine ions, and sits in a C_{2v} symmetry.

INTERPRETATION

Contributions of the second-order Doppler effect¹⁵ and interference effects¹⁶ due to dispersion terms in $E2$ transitions to the observed isomer

TABLE II. Mössbauer results with the 93.2-keV γ ray from Hf¹⁷⁸. Column 1: absorbers used in the experiments; column 2: thickness of absorbers in mg/cm² of Hf; column 3: observed FWHM in mm/sec; column 4: isomer shift observed with respect to W¹⁷⁸ source in Ta metal in mm/sec.

Absorber	d (mg/cm ² Hf)	Γ observed (mm/sec)	Isomer shift (mm/sec)
HfC	37	2.42 ± 0.09	-0.05 ± 0.03
Hf metal	89	2.75 ± 0.10	$+0.02 \pm 0.04^a$
HfO ₂	104	2.73 ± 0.10	$+0.03 \pm 0.04^b$ $+0.04 \pm 0.04^a$
Hf(Cp) ₂ Cl ₂	255	2.60 ± 0.18	$+0.21 \pm 0.05$

^a Spectrum fit to $\eta = 0$ constraining the intensities of the quadrupole triplet at 1 : 2 : 2.

^b Spectrum fit to $\eta \neq 0$ constraining the intensities of the 5 quadrupole components to be equal.

shifts deserve some comments. Following Ref. 16, on account of the dispersion terms, one may expect the isomer shift between the source and absorber to change. For the even-even heavy nuclei in this region, Wagner *et al.*¹⁶ have shown that, on account of dispersion terms, one may expect the center of gravity of a single line resonance to shift by ~ 0.04 mm/sec. It has been pointed out that, while comparing the isomer shifts between a pair of absorbers having the same effective thickness, (which is what we have done in the present work), such shifts will cancel. Conclusions of the present experiments hinge primarily on the isomer shift between absorbers of Hf(Cp)₂Cl₂ and Hf metal. On account of the differences in the Debye temperatures of these two absorbers, one may expect a second-order Doppler shift to be present. We have estimated this shift to be less than 0.02 mm/sec. In view of the comparatively large experimental errors on the observed shifts, in what follows, we have ignored these effects.

Recent measurements² of the isomer shift in Ru, Os, and Ir isotopes by the Mössbauer technique have revealed that the "s" electron density in chemical compounds of these elements (where these 4d and 5d transition elements occur in different oxidation states) shows the same trends as in iron, a well-studied 3d transition element. In particular, the s electron density at the metal ion in the organometallic ferrocene, ruthenocene, and osmocene, is in general smaller than the s electron density in the corresponding metal lattice. The two cyclopentadienyl rings contribute 10 electrons to the molecular bond with the metal ion and it is the occupancy of the d_{z^2} , $d_{x^2-y^2}$, and d_{xy} molecular orbitals located mainly on the metal ion that shields the s electrons from the nucleus and thereby lowers the s electron density. One would expect a similar situation to exist when one considers the s electron density at the hafnium nucleus in Hf(Cp)₂Cl₂ and hafnium metal. The positive sign of the isomer shift between Hf(Cp)₂Cl₂ and Hf metal (Table I) should be interpreted to indicate that $\delta\langle r^2 \rangle$ for Hf¹⁷⁸ is negative.

In order to arrive at the value of $\delta\langle r^2 \rangle(\text{Hf}^{178})$, the shrinkage in the nuclear charge radius of Hf¹⁷⁸, it is necessary to make a quantitative estimate of $\Delta|\psi(0)|^2(\text{Hf})$, the change in the "s" electron density at hafnium, between Hf(Cp)₂Cl₂ and Hf metal. Recently, Kaindl *et al.*² have observed an isomer shift of $+2.05 \pm 0.25$ mm/sec between Os(Cp)₂ and Os metal in Mössbauer effect studies of the 36.3-keV γ ray in Os¹⁸⁹. Using their value for $\delta\langle r^2 \rangle(\text{Os}^{189}) = -2.1 \times 10^{-3} \text{ fm}^2$, we calculate $\Delta|\psi(0)|^2(\text{Os})$, the change in the "s" electron density between osmocene and Os metal, to be -4.98

$\times 10^{-26} \text{ cm}^{-3}$. In order to correlate $\Delta |\psi(0)|^2(\text{Hf})$ with $\Delta |\psi(0)|^2(\text{Os})$, we make the following assumptions: (a) The "s" electron density at Hf in $\text{Hf}(\text{Cp})_2\text{Cl}_2$ and Os in $\text{Os}(\text{Cp})_2$ are the same. (b) The s electron density at Hf in Hf metal ($5d^26s^2$) is larger than that at Os in Os metal ($5d^66s^2$) on account of the screening of the 6s charge density by the additional four 5d electrons in Os metal. Following Brix,¹⁰ we assume each 5d electron screens the 6s charge density by 20%. It then follows, as a result of these assumptions, that $\Delta |\psi(0)|^2(\text{Hf})$ is equal to 2.07 times $\Delta |\psi(0)|^2(\text{Os})$.

The procedure outlined above for estimating $\Delta |\psi(0)|^2(\text{Hf})$ suffers from some weaknesses. First, we have assumed that the electron states of Hf in Hf metal and Os in Os metal to be given by the free atom configurations. This may be examined in the light of some recent isomer shift results^{2, 3} that suggest the electron states of Os and W in the corresponding metal structures to be described by Os(VI) $5d^2$ and W(V) $5d^1$ configurations. It would then appear very likely that the case of Hf in Hf metal corresponds to a Hf(IV) $5d^0$ configuration. This would also explain the smallness of the observed isomer shift¹ between Hf metal and HfO_2 . In an attempt to correlate the s electron density at Hf in Hf metal with that at Os in Os metal, it would appear more reasonable to consider shielding by two 5d electrons rather than four. Secondly, the density of Hf metal (13.29 g/cm^3) is smaller than the density of Os metal (22.57 g/cm^3). Both these considerations suggest that assumption (b) somewhat overestimates the change in the s electron density $\Delta |\psi(0)|^2(\text{Hf})$. From the above considerations a more realistic estimate of $\Delta |\psi(0)|^2(\text{Hf})$ would be to enhance $\Delta |\psi(0)|^2(\text{Os})$ by a factor of 1.44 to correspond to the shielding of two 5d electrons. This then leads to

$$\Delta |\psi(0)|^2(\text{Hf}) = -7.2 \times 10^{-26} \text{ cm}^{-3},$$

$$\delta \langle r^2 \rangle (\text{Hf}^{178}) = -0.37 \times 10^{-3} \text{ fm}^2,$$

$$\delta \langle r^2 \rangle / \langle r^2 \rangle (\text{Hf}^{178}) = -0.13 \times 10^{-4}.$$

DISCUSSION

The present result on Hf^{178} is similar to recent Mössbauer-isomer-shift results on W^{182} ³ and Yb^{174} ,¹⁷⁶⁴ where a shrinking (negative $\delta \langle r^2 \rangle$) of the nuclear charge radius in the first 2^+ excited state of these deformed nuclei has been inferred (Table III). The magnitudes of these shifts are small and in fact, in all these studies, the largest isomer shift observed is about 10% of the natural line-width. The experimental procedure, the choice of absorbers employed, the stability of absorbers

with respect to their chemical state of oxidation, and the use of sources having narrowest emission linewidths then become important considerations in these experiments. It is therefore not surprising that there have been Mössbauer isomer shift measurements on these very isotopes, viz., W^{182} , Yb^{174} , and Hf^{178} ,¹⁸⁰ that have yielded just the opposite result, viz., an increase (positive $\delta \langle r^2 \rangle$) of the nuclear charge radius in the 2^+ state (Table IV). The positive value of $\delta \langle r^2 \rangle$ for Hf^{178} by Snyder, Ross, and Bunbury¹ was based on the isomer shift observation between Hf metal and HfO_2 . Both these hafnium hosts are noncubic and show a partially resolved quadrupole splitting. Extraction of a small and reliable isomer shift from these data is difficult. We have repeated these experiments. A careful analysis of the data has shown no evidence of a measurable isomer shift between Hf metal and HfO_2 . These results confirm the suggestion made earlier that from the standpoint of isomer shifts, both Hf metal and HfO_2 may represent a case of Hf in the tetravalent state. Unfortunately, unlike Os, Hf does not form many stable chemical compounds possessing different states of oxidation, thus forcing the choice of an organometallic compound for isomer shift studies. Organometallic compounds are not very suitable for Mössbauer measurements on account of their characteristic low Debye temperatures. Furthermore, a quantitative analysis of the isomer shifts in organometallic compounds does become involved on account of the molecular orbital theory approach. Yet their use in Mössbauer isomer shift studies, whenever possible, has some advantages. It is well known that Fe^{57} , Ru^{99} , and Os^{189} show the largest isomer shift in their di-cyclopentadienyl derivative. Also it is possible while using organometallic compounds, to predict in many cases the sign of $\Delta |\psi(0)|^2$. Our use of $\text{Hf}(\text{Cp})_2\text{Cl}_2$ in Mössbauer studies with an even isotope clearly shows the negative sign of $\delta \langle r^2 \rangle$ for Hf^{178} . We have recently performed Mössbauer measurements with Hf^{180} ¹⁷ and have observed a shrinking of the same magnitude in the nuclear charge radius of Hf^{180} in the 2^+ state. The magnitude of the isomer shift observed in the present experiment may be compared with other such measurements that have suggested a negative $\delta \langle r^2 \rangle$ in Table III. In our judgment the present experiments present the clearest evidence of a shrinking of the nuclear charge radius in the first 2^+ rotational state.

This is a strange result if one naïvely thinks that the charge radius and mass radius both should stretch in the higher rotational states. Recent experimental results^{18, 19} indicate that the neutron radius in the ground state of atomic

TABLE III. Summary of Mössbauer results that show a shrinking of the nuclear charge radius in the first rotational state of even-even nuclei.

Isotope	$\delta \langle r^2 \rangle$ (10^{-3} fm^2)	$\delta \langle r^2 \rangle / \langle r^2 \rangle^a$ (10^{-4})	Isomer shift observed	Reference
W ¹⁸²	-0.44	-0.16	WCl ₆ - (WCl ₆) ²⁻ = -0.07 mm/sec	3
Yb ¹⁷⁴	-0.54 ± 0.19	-0.20 ± 0.07	Yb ³⁺ - Yb ²⁺ = -0.132 ± 0.023 mm/sec	4
Yb ¹⁷⁶	-0.19 ± 0.09	-0.07 ± 0.03	Yb ³⁺ - Yb ²⁺ = -0.047 ± 0.016 mm/sec	4
Hf ¹⁷⁸	-0.37	-0.13	Hf metal - Hf(Cp) ₂ Cl ₂ = -0.21 ± 0.06 mm/sec	Present

^a The $\langle r^2 \rangle$ values are computed using the formula $\langle r^2 \rangle = \frac{2}{5}[1.123A^{1/3} + 2.352A^{-1/3} - 2.070A^{-1}]^2$ from L. R. B. Elton, *Nuclear Sizes* (Oxford U. P., Oxford, England, 1961).

TABLE IV. Summary of Mossbauer results on isotopes listed in Table III, that show an increase of the nuclear charge radius in the first rotational state.

Isotope	$\delta \langle r^2 \rangle$ (10^{-3} fm^2)	$\delta \langle r^2 \rangle / \langle r^2 \rangle$ (10^{-4})	Isomer shift observed	Reference
W ¹⁸²	+3.4	+1.3	W metal - WCl ₆ = +0.25 mm/sec	a
Yb ¹⁷⁴	+0.7	+0.27	Yb ³⁺ - Yb ²⁺ = +0.16 mm/sec	b
Hf ¹⁷⁸	+0.7	+0.26	Hf Metal - HfO ₂ = +0.09 mm/sec	1
Hf ¹⁸⁰	+0.8	+0.29	Hf metal - HfO ₂ = +0.10 mm/sec	1

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nuclei is larger than the proton radius. In the higher rotational states, one would expect the mass radius to stretch more than the charge radius. What causes the charge radius to contract in the higher rotational states is not at all clear. It may be pointed out that Scharff-Goldhaber and Goldhaber²⁰ infer a reduction in the value of the transition quadrupole moment of the higher rotational states in some hard even-even rotors.

states in some hard even-even rotors.

It is important to confirm by Mössbauer technique the negative isomer shifts observed by muonic x-ray studies and to make a systematic study to recognize a pattern in the behavior of the nucleus.

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PHYSICAL REVIEW C

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Electromagnetic Properties of Even-Even Platinum Nuclei

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The lifetimes of the first and second excited states were determined in ^{194,196}Pt with delayed coincidence techniques: ¹⁹⁴Pt $\tau(2^+) = 50 \pm 5$ psec, $\tau(2'^+) = 51 \pm 6$ psec, ¹⁹⁶Pt $\tau(2^+) = 43.6 \pm 3.0$ psec, $\tau(2'^+) = 52 \pm 5$ psec. For this latter level the crossover/stopover intensity ratio is $\leq 6.10^{-5}$. Deduced $B(E2)$ transition probabilities show a simultaneously diminishing trend for $2'^+ \rightarrow 0^+$ and $2^+ \rightarrow 2^+$ versus atomic mass number. New lifetimes are used to reevaluate nuclear g factors, which all lie around $g_{2^+} \approx 0.3$. Phenomenological anharmonic vibration-model calculations reproduce well the measured quadrupole transition probabilities and nuclear g factors.

I. INTRODUCTION

The even-even platinum isotopes occupy a remarkable place among the transition nuclei. The presence of $2'^+$ states at an energy twice that of 2^+ states and the equality of g_{2^+} and $g_{2'^+}$ factors for each isotope suggest that platinum isotopes should be at the edge of the nearly harmonic nuclei. The existence of large static quadrupole moments for 2^+ states shows that appreciable deviations from harmonicity are present.¹ Another test of the validity of the harmonic picture is the magnitude of the ($2'^+ \rightarrow 0^+$) crossover transition ($\Delta N = \pm 2$).

This paper is the continuation of a systematic study of electromagnetic properties of even-even platinum nuclei.^{2,3}

II. EXPERIMENTS

A. Lifetimes

Lifetimes have been measured by delayed coincidences between conversion electrons and γ or x

rays. Conversion electrons were selected with a lense-type β spectrometer. γ and x rays were detected with a plastic scintillator XP1021 photomultiplier assembly. To avoid transit time variations of electrons of different energies in the spectrometer, the magnetic field was kept constant and an electric preacceleration varied electron energy.⁴ Coincidences were taken with a conventional fast-slow setup. To cancel drifts of time correlation curves versus energy, a compensation unit has been coupled to the time-to-pulse-height converter.⁵ The compensation was set with the continuous spectrum of a ⁶⁰Co source for each measurement on the energy range to be compensated. To avoid shifts due to source intensity or temperature variations, we performed short runs at each point.

1. 2^+ and $2'^+$ Levels in ¹⁹⁴Pt

Figure 1 shows the simplified scheme of the ¹⁹⁴Ir β decay.⁶ ¹⁹³Ir enriched to 95% has been irradiated in a reactor to form ¹⁹⁴Ir (24-h) activity. The elec-