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Beta decay of ^{187}Re and cosmochronology

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Uncertainties which limit the use of the ^{187}Re-^{187}Os isobaric pair as a cosmochronometer for the age of the galaxy and the universe include those of the partial half-lives of the continuum and bound-state decays of ^{187}Re. While the total half-life of the decay is well established, the partial half-life for the continuum decay is uncertain, and several previous measurements are not compatible with each other. A high-temperature quartz proportional counter has been used in this work to remeasure the continuum decay of ^{187}Re by introducing a metallo-organic rhenium compound into the counting gas. The measured beta end-point energy for the continuum decay of neutral ^{187}Re to singly ionized ^{187}Os of 2.70 ± 0.09 keV agrees with earlier results. However, the present half-life measurement of (45 ± 3) Gyr agrees within the quoted uncertainties only with an earlier measurement. The new half-life for the continuum decay and the recently reported total half-life of (43.5 ± 1.3) Gyr yield a branching ratio for the bound-state decay into discrete atomic states of (3 ± 6) %. This is in agreement with the most recent calculated theoretical branching ratio of approximately 1%.

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I. INTRODUCTION

In 1964 Clayton [1] introduced the Re-Os isobaric pair as an absolute cosmochronometer. This cosmochronometer is of special interest due to the long half-life of ^{187}Re which is comparable to the known age of the Universe. The cosmochronometer is discussed briefly in Sec. V. Although there is general agreement for the measured total half-life of ^{187}Re, the partial half-lives for the continuum and bound-state decays are still uncertain. The experimental half-lives of ^{187}Re measured since 1962 are listed in Table I. The various determinations for the total half-lives based on direct and indirect methods are in good agreement. The most recent determination of the total half-life [2] was obtained directly from the buildup of ^{187}Os in a rhenium sample over a few years. With one exception, all determinations of the half-life for continuum decay were carried out with gas counters. As seen from the measurements included in Table I, there exist obvious discrepancies between the measured partial half-lives. Compatibility within the reported uncertainties can only be achieved at the 2σ level.

^{187}Re with its unique first-forbidden beta decay has a long half-life (45 Gyr) and the lowest β endpoint energy known in nature (2.6 keV). This severely limits the choice of detector used in the determination of the spectral shape and half-life of ^{187}Re. Detectors with an entrance window are not suitable for the detection of such low-energy electrons. Furthermore, the use of solid sources of rhenium results in self-absorption and corrections are difficult. Attempts at using solid rhenium samples, as reported by Watt and Glover [3] and others, have not produced satifactory results.

Proportional counters with internal gaseous sources are the best candidates for the study of the decay of ^{187}Re. Here, metallo-organic vapors containing natural rhenium (which contains 62.6% of ^{187}Re) are added to the counting gas, often P-10 (90% argon, 10% methane). Elevated temperatures are usually needed to ensure sufficient vapor pressures. Although solid-state detectors could in principle be doped with low concentrations of rhenium, the long half-life of rhenium would require large-area and/or large-volume detectors. Due to the necessarily low concentrations of rhenium in such solid-state detectors there would also be problems with natural background. Gas scintillation counters have better energy resolution than proportional counters, but they are not 100% efficient. Likewise, the use of liquid scintillation counters requires knowledge of the detector efficieny at low energies. Naldrett [4] has recently used a liquid scintillation counter doped with the organic rhenium compound [$ReOCl_3{\cdot}2P(C_6H_5)_3$]. The author reported a partial half-life which is shorter than the accepted value for the total half-life. This contradiction is likely due to uncertainties in the detector efficiency when extrapolated to very low energies [4].

High-temperature proportional counters have previously been used for the measurement of the spectral shape and half-life of ^{187}Re. The major problem in the use of high-temperature proportional counters consists in finding an appropriate rhenium compound that is volatile and yet is not a counter poison. Biscyclopentadienylrhenium hydride (ReCPD) is the only rhenium compound that has been successfully used [5–7]. However, decomposition of the compound has been observed in metal counters. The long half-life of 66 Gyr reported by Brodzinski and Conway [5] can probably be attributed in part to this

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TABLE I. Measurements of the half-life of ^{187}Re since 1962.

$T_{\frac{1}{2}}$ (Gyr)	$E_{\max}$ (keV)	Method	Reference
		Total decay	
43 ± 5		Indirect; molybdenites of known age	Hirt *et al.*, 1962; Ref. [37]
42.8 ± 2.1		Indirect; meteorites of known age	Luck, Birck, and Allegre, 1980; Ref. [38]
45.6 ± 1.2		Indirect; meteorites of known age	Luck and Allegre, 1983; Ref. [39]
43.5 ± 1.3	–	Direct; growth of ^{187}Os	Linder *et al.*, 1986; Ref. [2]
		Continuum decay	
120 ± 40	~3	Geiger counter (ReO_3Cl vapor)	Wolf and Johnston, 1962; Ref. [40]
~30	1.2 ± 0.1	Proportional counter (solid sample)	Watt and Glover, 1962; Ref. [3]
66 ± 13	2.62 ± 0.09	Proportional counter (ReCPD vapor)	Brodzinski and Conway, 1965; Ref. [5]
47 ± 5	2.54 ± 0.03	Proportional counter (ReCPD vapor)	Payne and Drever, 1965; Ref. [6]
	2.65 ± 0.04	Proportional counter (ReCPD vapor)	Huster and Verbeek, 1967; Ref. [7]
35.1 ± 3.7		Liquid scintillation counter [$ReOCl_3{\cdot}2P(C_6H_5)_3$]	Naldrett, 1984; Ref. [4]
45 ± 3	2.70 ± 0.09	Proportional counter (ReCPD vapor)	Present work, 1992

problem. Specifically, due to the partial decomposition of ReCPD, an excess amount of ReCPD was introduced by the authors into the metal counter, and the amount of ReCPD in the active volume was estimated using a vapor pressure equation. In this investigation a substantial broadening of the ^{37}Ar calibration peak was also observed in the presence of ReCPD.

II. EXPERIMENTAL TECHNIQUES

The successful use of radioactive $Ho(fod)_3$ in a stainless-steel proportional counter in the determination of the M and N electron-capture ratio of ^{163}Ho by Hartmann and Naumann [8] led us to investigate the feasibility of the synthesis of beta-diketonate compounds [9] of rhenium for use as additive to the counting gas. These compounds are generally stable and highly volatile at relatively low temperatures. The large number of metal (Me) beta-diketonates can be classified depending upon the mode of bonding of ligand moieties to metal atoms. One class of metal beta-diketonates includes $Me(fod_3$, $Me(thd)_3$, $Me(fac)_3$, and $Me(acac)_3$. Here, the central metal ion is oxygen bonded to three bulky groups forming a neutral atom.

A multiwire wall-less stainless-steel proportional counter with an active volume of about 5000 cm^3 and a total volume of about 10 000 cm^3 was originally designed and constructed [10] for the measurement of the partial half-life of ^{187}Re. Fused quartz tubes (1–3 mm thickness) were initially used for electrical insulation inside the counter. Although the detector performed satisfactorally at room temperature, spurious signals were observed at elevated temperatures. These spurious signals are believed to be due to electrical discharges at the heated quartz insulation. Isozumi *et al.* [11] reported similar problems with quartz insulators. Subsequently, teflon tubes were used for insulation and they performed satisfactorily at elevated temperatures.

The response of the detector in the presence of $Ho(fod)_3$ or $Sm(thd)_3$ was investigated by introducing 30–100 mg of these compounds into the detector. At elevated temperatures the presence of both compounds resulted in a gradual decrease in gain and a broadening of the 5.9 keV ^{55}Fe calibration peak. This behavior may be due to a reaction between the compounds and the inner metal surfaces of the counter, resulting in the decomposition of the compounds into a counter poison. Alternatively, these compounds themselves could in fact be counter poisons. This problem was not reported by Hartmann [8], though, possibly because the concentration of $Ho(fod)_3$ in their investigation was significantly lower than the concentrations used in the present study. The contamination of our detector by some compound that acted as a catalyst in a reaction with $Ho(fod)_3$ and $Sm(thd)_3$ is not considered likely, because the detector was baked several times at temperatures of over 300 °C.

Attempts to synthesize $Re(fod)_3$ resulted in only small yields [10] which were not sufficient to allow their use in the counter. $Re(fod)_3$ was prepared by a method similar to the procedure used by Sievers and Connolly [12] to synthesize $Mn(fod)_3$. $Re(thd)_3$ was prepared by a method similar to the procedure used to synthesize $Re(fac)_3$ [13]. The latter compound was contaminated by triphenylphosphine, but due to the small yield no attempts were made to purify the compound.

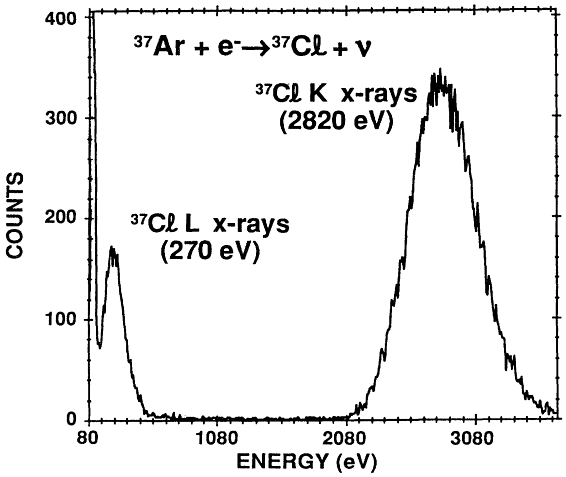

FIG. 1. Energy spectrum for *K* and *L* capture x rays from an ^{37}Ar source obtained with a quartz counter at 180 °C.

The problems encountered with the stainless-steel counter and the use of beta-diketonate compounds led to the decision to build a quartz counter and to revert to the use of ReCPD. This metallo-organic compound was synthesized using the procedures reported by Green *et al.* [14]. A reduced temperature of 180 °C was employed to minimize decomposition even though ReCPD has been reported thermally stable to at least 250 °C [4]. The quartz counter had no exposed inner metal surfaces and was designed and constructed to overcome the problems with counter poisoning. This counter was indeed successfully used in the determination of the half-life and the spectral shape of ^{187}Re in the present investigation. The detector response was investigated by adding traces of radioactive ^{37}Ar gas to the P-10 counter gas. An ^{37}Ar spectrum is shown in Fig. 1. The relative energy resolution of the 2.82 keV chlorine *K* x-ray line is about 24% full width at half maximum (FWHM). The 270 eV chlorine *L* x-ray line is also well resolved but resides on low-energy noise which extends to 300–400 eV. Spectra for ^{187}Re without and with traces of ^{37}Ar are shown in Figs. 2 and 3. The background has a broad maximum at about 5 keV and is identified as mostly due to cosmic ray muons. It is shown in Fig. 4. The resolution of the 2.82 keV chlorine x-ray line in the presence of ReCPD is 25% (FWHM). The introduction of ReCPD resulted in an increase in gain. This increase can be accounted for if ReCPD has a smaller ionization potential than argon. A decrease in count rate by about 3% over 24 h was observed in this experiment. It can be attributed to the diffusion of ReCPD into small capillaries. A similar effect was observed by Payne [6].

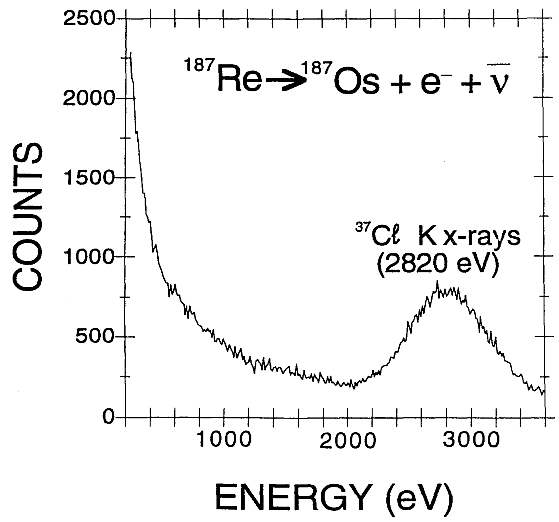

FIG. 3. Energy spectrum for ^{187}Re with background and traces of ^{37}Ar (for energy calibration) obtained with a quartz counter at 180 °C.

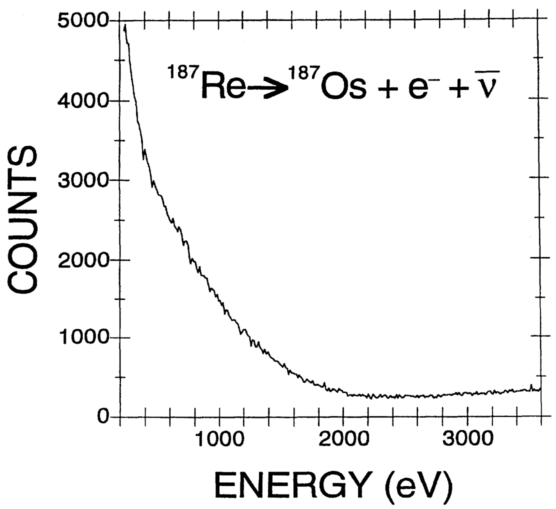

FIG. 2. Energy spectrum for the beta decay of ^{187}Re including background obtained with a quartz counter at 180 °C.

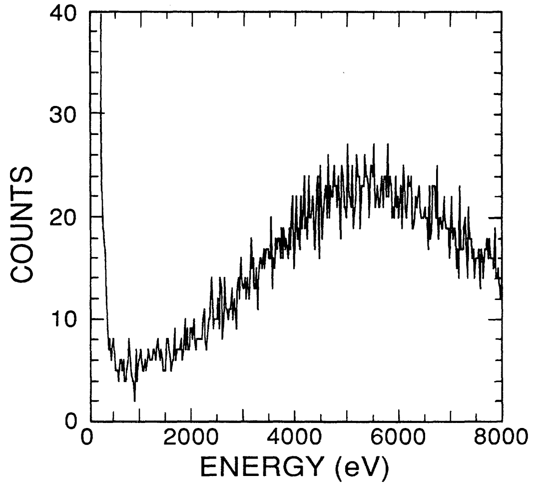

FIG. 4. Background energy spectrum obtained with a quartz counter at 180 °C.

III. SPECTRAL SHAPE OF THE BETA DECAY OF ^{187}Re

The theoretical spectral shape of the continuum first-forbidden unique transition of $^{187}\mathrm{Re} \longrightarrow {}^{187}\mathrm{Os} + e^- + \bar{\nu}$ with $J^\pi = 5/2^+ \longrightarrow J^\pi = 1/2^-$ is

$$\frac{dN\,(W)}{dW} = A\,C\,W\,(W_0 - W)\,p\,\sqrt{(W_0 - W)^2 - m_{\bar{\nu}}^2} \tag{1}$$

with

$$C = F_0\,L_0\,(W_0 - W)^2 + \frac{9}{2R^2p^2}\left[g_{-2}^2 + f_{-2}^2\right]_{r=R}\ . \tag{2}$$

Here, A is a normalization constant, W is the total energy, W_0 is the end-point energy, p is the electron momentum, $m_{\bar{\nu}}$ is the antineutrino mass, $(F_0\,L_0)$ is the Fermi fuction, and R is the nuclear radius. The expression for the spectral shape also contains the Dirac electron radial wave funtions $g(r)$ and $f(r)$ evaluated at the nuclear surface. There are no known analytical solutions for the general potential where the effects of the finite nuclear size and screening are included. The radial wave functions can be obtained for the general case by solving the Dirac equations numerically [15,16]. The solution to the Dirac equations is given by Rose [17] assuming a point nucleus charge distribution without screening and $V(r) = -Z\,e^2/r$. Gove and Martin [18] obtained similar equations for low electron energies. The latter equations were used in the present work.

The effect of screening has been considered by Rose [19], and approximate analytical expressions describing screening corrections are available. However, these expressions are not valid at very low energies. Behrens and Jänecke [20] published an extensive tabulation of the various relevant parameters which include the effects of screening and of the finite nuclear size. However, the published tables do not include the parameters for very low energies needed in the analysis of the ^{187}Re spectra. The screening corrections are especially important for high-Z, low-energy transitions [15,16]. A study of the above-mentioned tables indicates that the screening corrections for L_0 are approximately constant at low energies, but the value of the screening corrections for the second term in Eq. (2) changes significantly at low energies. In the analysis of our ^{187}Re spectra an empirical energy-dependent term was therefore introduced to account for the screening corrections. Although the inclusion of this term did not significantly affect the general spectral shape, it does have a noticeable effect on the endpoint energy.

The influence of the finite nuclear size has been treated by Rose and Holmes [21]. The corrections are estimated to be less than 1% for ^{187}Re, and they were not included in the analysis of the present spectra.

IV. EXPERIMENTAL RESULTS

The theoretical spectrum was folded with the response function extracted from the ^{37}Ar spectrum assuming a $W^{-\frac{1}{2}}$ energy dependence for the experimental linewidth. The experimental data were fitted to the folded theoretical spectrum using a non-linear least-squares fit procedure contained in the computer program NucUM [10]. Background from an independently measured spectrum, renormalized in gain and intensity, was subtracted from the measured ^{187}Re spectrum. The resulting fit is shown in Fig. 5. Below 400 eV the data points are higher than the calculated curve due to the onset of random electronic noise. A small excess of events is observed at high energies due to residual background. However, it was felt that a purely empirical background subtraction was not justified. The weak effect on the extracted spectrum parameters was estimated and is included in the results. A value of 2.70 ±0.09 keV was obtained for the ^{187}Re endpoint energy. This value is slightly higher than the endpoint energies reported by other authors (see Table I). Brodzinski and Conway [5] and Huster and Verbeek [7] used a simplified form for the theoretical spectral shape in the analysis of their data. Payne [6], on the other hand, used the complete expression, included the effects of finite nuclear size but did not consider screening corrections. An analysis of our data without the empirical energy-dependent term, which was introduced to account for the screening corrections, lowers the end-point energy. This might explain the relatively low value reported by Payne [6].

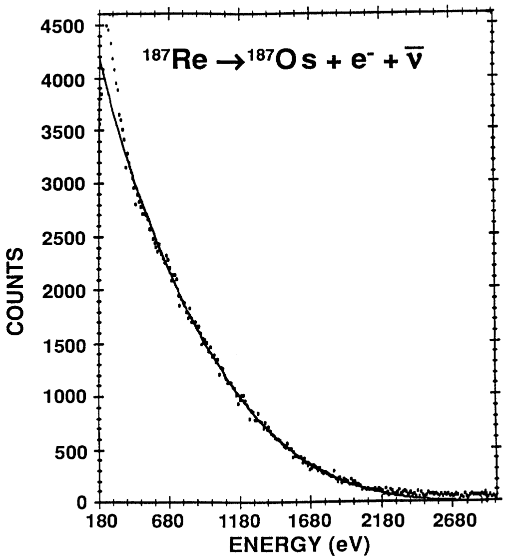

FIG. 5. Corrected energy spectrum for the β^- decay of ^{187}Re including a calculated fit (see text).

The extrapolated spectrum was used to determine the partial half-life of ^{187}Re. A mass of 22.6 ± 0.9 mg of ReCPD was used in the present measurement of the spectral shape of the decay of ^{187}Re. The molecular weight of ReCPD was taken as 317.5, and the 63% natural abundance of ^{187}Re was assumed. The active length of the detector of 68.8 ± 1.0 cm is defined by the field correction tubes. The total volume of the detector was obtained by sharing various amounts of P-10 gas with a known volume and observing the change in pressure. Uncertainties in the spectral fitting procedure include the results from fitting over different ranges of energies and the extrapolation of the theoretical spectrum to low energies. A decay rate of (13.1 ± 0.6) events per second was obtained from the extrapolated fitted spectrum.

A partial half-life of the continuum β decay of ^{187}Re to singly ionized ^{187}Os of 45 ± 3 Gyr is extracted from the measurements. Based on this half-life for the continuum decay and a total half-life of (43.5 ± 1.3) Gyr reported by Linder *et al.* [2], the branching ratio for the bound-state decay into discrete atomic states is estimated to be (3 ± 6)%.

A theoretical ratio of bound state to continuum decay of 58% (or 37% for the ratio bound state to total decay) has been calculated earlier by Perrone [22] in agreement with the experimental ratio of 50 ± 30% deduced from the partial half-life of 66 ± 13 Gyr reported by Brodzinski and Conway [5]. Nonrelativistic hydrogenlike wave functions were used in this calculation [22], and exchange and overlap effects were neglected. However, two more recent calculations yield much smaller ratios. Williams *et al.* [23] obtained a ratio of approximately 1% using Dirac wave functions and a modified Lenz-Jensen potential. A ratio of approximately 1% was also obtained by Chen *et al.* [24] in similar calculations using the Hartee-Fock-Dirac approximation. The branching ratio of (3 ± 6)% reported in the present work is in agreement with these latter calculations, and it is concluded that the β decay of neutral ^{187}Re is strongly dominated by the decay into the continuum.

V. RHENIUM COSMOCHRONOLOGY

Cosmochronology (e.g., Ref. [25]) encompasses determinations (i) of the age of the Universe, mostly from the measurement of the Hubble constant, (ii) of the age of our Galaxy, mostly from the age of the oldest stars in globular clusters, and (iii) the age of the chemical elements as deduced from radioactive isotopes, their production in the Galaxy prior to the formation of the solar system, and their decay afterwards.

Following the condensation of the solar material shortly after its formation, radioactive decays, e.g., $^{87}\mathrm{Rb} \rightarrow {}^{87}\mathrm{Sr}$, changed the relative abundance of parent and daughter nuclei. Using several such decay processes, the oldest materials from the Earth, moon, and meteorites (geochronology) are dated at 4.6 Gyr.

Similarly, cosmochronology (nucleo-cosmochronology) relates relative isotopic abundances to radioactive decays, but in addition knowledge about nucleosynthesis processes is required, primarily about the *r* process during supernovae explosions and about the *s* process. The selection of *r* process pairs of nuclei with appropriate life times, e.g., ^{232}Th-^{238}U, ^{235}U-^{238}U, ^{244}Pu-^{238}U, or other pairs including ^{187}Re-^{187}Os has led to powerful cosmochronometers. These were reviewed in detail very recently [30].

The Re-Os isobaric pair was introduced by Clayton [1] as an absolute *r* process cosmochronometer. Its features were discussed extensively by Symbalisty and Schramm [26]. As noted, the long half-life of ^{187}Re is appropriate for a cosmochronometer probing the age of the Galaxy and the Universe. The use of the ^{187}Re-^{187}Os abundance ratio to estimate the duration of nucleosynthesis has the advantage that it is not affected by uncertainties in the *r* process production rate of ^{187}Re. The nucleus ^{187}Re is generally classified as a pure *r* process isotope since it is bypassed in the *s* process by the β decay of ^{186}Re. In contrast, the nuclei ^{186}Os and ^{187}Os are pure *s*-process isotopes. They are shielded from the *r* process by ^{186}W and ^{187}Re, respectively. It is in principle possible to determine the amount of radiogenic ^{187}Os by subtracting the *s* process contributions and then estimate the duration of nucleosynthesis. However, there still exist several complications which limit the use of the ^{187}Re-^{187}Os isobaric pair as a cosmochronometer for the age of the Galaxy and the Universe. These effects will be discussed briefly below.

One major uncertainty in the use of the ^{187}Re-^{187}Os pair as cosmochronometer are the chronological models used for describing the nucleosynthesis of heavy elements prior to the formation of the solar system. The rate of nucleosynthesis of heavy elements since the formation of galaxies is not well known. There are in existence several chronological models that assume either uniform, or exponential, or sudden synthesis [25].

Furthermore, the half-life of ^{187}Re depends strongly on its degree of ionization [27,28] and hence the temperature. Continuum decay for neutral ^{187}Re is energetically just barely possible because the negative nuclear energy is compensated by the increased atomic energy (due to the increase in nuclear charge). Continuum decay is energetically not possible for highly ionized ^{187}Re. Bound-state decay into empty atomic orbits, on the other hand, is increasingly favored with increasing ionization. The total half-life of ^{187}Re therefore becomes strongly temperature dependent [29]. The unique first-forbidden continuum decay $^{187}\mathrm{Re} \longrightarrow {}^{187}\mathrm{Os}$ with $J^\pi = 5/2^+ \longrightarrow J^\pi = 1/2^-$ dominates at low temperatures, but the decay of highly ionized ^{187}Re is dominated by the bound-state decay. At the highest ionization, including fully ionized rhenium, the unique first-forbidden bound-state decay of ^{187}Re with $J^\pi = 5/2^+$ to the ground state of ^{187}Os with $J^\pi = 1/2^-$ is replaced by the much faster nonunique first-forbidden bound-state decay to the first excited state of ^{187}Os at $E_x = 9.75$ keV with $J^\pi = 3/2^-$. As a consequence the half-life of ^{187}Re is reduced under stellar conditions by several orders of magnitude. After ^{187}Re is synthesized, subsequent reprocessing through the interior of stars at high temperatures lowers the effective half-life. The degree and the conditions of recycling of rhenium may significantly reduce the estimated age of the Galaxy based on the ^{187}Re-^{187}Os isobaric pair. Yokoi, Takahashi, and Arnould [29] reported compatibility with an age of the Galaxy in the approximate 11–15 Gyr range in agreement with the chronometric age predictions for the galaxy reported in a recent review [30] which lie mostly in the range 12–15 Gyr.

It is interesting to note that newly developed techniques [31] have recently been used to measure the bound-state β^-decay of bare ^{163}Dy^{66+} ions using a heavy-ion storage ring. There is very good agreement with a calculated value [28]. A direct measurement of the bound-state decay of fully ionized ^{187}Re to the ground and first-excited state of ^{187}Os has thus become feasible for future comparison with the theoretical prediction of the absolute decay rate [22,28] which is estimated to be uncertain by a factor of 2. The calculated enhancement of the nonunique first-forbidden decay over the unique first-forbidden decay by 3–5 orders of magnitude appears high but may be explained by a hindrance introduced in the decay to the ground state due to the asymptotic Nilsson quantum numbers. An experimental confirmation of the theoretical predictions, which serve as a basis for present nucleochronology calculations, is therefore desirable.

As emphasized by many authors (e.g., [29,30]), various other effects introduce uncertainties into the use of the ^{187}Re-^{187}Os chronometer. Under r process conditions immediately after the synthesis of ^{187}Re when the temperatures are in the range of several 100 keV, even ^{187}Re can occupy excited states. Other decay channels become accessible (e.g., $J^\pi=7/2^+ \longrightarrow J^\pi=3/2^-$) which may result in an added reduction in half-life. These conditions, however, last only for a short time, and the contributions to the relative abundance of ^{187}Re and ^{187}Os are estimated to be minimal.

Another problem arises due to the temperature dependence of the neutron capture cross section of ^{187}Os needed to estimate the s process abundances. Under stellar conditions, i.e., at temperatures equivalent to approximately 30 keV, ^{187}Os nuclei occupy the low-lying excited state at 9.75 keV, whereas the measured neutron capture cross section is that of ^{187}Os in its ground state. Neutron capture cross section calculations for the low-lying excited states of Os have been made [32,33]. A correction factor, F, was introduced to account for the difference between laboratory and stellar cross section ratios:

$$\langle\sigma^*_{186}\rangle \Big/ \langle\sigma^*_{187}\rangle = F \, \langle\sigma_{186}\rangle \Big/ \langle\sigma_{187}\rangle . \tag{3}$$

Here, $\langle\sigma^*\rangle$ is a Maxwell-Boltzmann-averaged stellar cross section for a thermally populated distribution of excited states, and $\langle\sigma\rangle$ is the corresponding average cross section for the ground state. Initial estimates [32,33] of $0.8 \leq F \leq 1.1$ were later refined [34,35] on the basis of neutron elastic and inelastic scattering data, measurements of neutron capture cross sections for ^{189}Os, which has the same Nilsson single-quasiparticle quantum numbers as the first excited state of ^{187}Os, and Hauser-Feshbach compound nucleus calculations. The new value of $F = 0.82\pm0.02$ essentially removes the uncertainty from the temperature dependence of the ^{187}Os neutron capture cross section.

Other problems with the ^{187}Re-^{187}Os pair are possible s-process branchings in the W–Os region [36] and also ^{187}Os electron capture during astration [29].

VI. SUMMARY

A quartz proportional counter was used to remeasure the partial half-life of the continuum decay of neutral ^{187}Re into singly ionized ^{187}Os by adding the metallo-organic compound ReCPD to the counting gas. The spectrum of the unique first-forbidden β decay of ^{187}Re was found in good agreement with the theoretically predicted spectral shape with a β^- end-point energy of 2.70 ± 0.09 keV. The measured partial continuum half-life for the decay of 45 ±3 Gyr is in agreement with an earlier unpublished measurement [6] but contradicts several published values. The resulting branching ratio for the decay of ^{187}Re into bound states of ^{187}Os of $(3 \pm 6)\%$ is in agreement with the recent calculations of Williams *et al.* [23] and Chen *et al.* [24].

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