VOLUME 77, NUMBER 26 PHYSICAL REVIEW LETTERS 23 DECEMBER 1996

Observation of Bound-State β^- Decay of Fully Ionized ^{187}Re: ^{187}Re-^{187}Os Cosmochronometry

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(Received 20 September 1996)

We observed the bound-state β^- decay of fully ionized ^{187}Re nuclei circulating in a storage ring. With two independent methods the time dependent growth of hydrogenlike ^{187}Os ions has been measured and a half-life of 32.9 ± 2.0 yr for bare ^{187}Re nuclei could be determined, to be compared with 42 Gyr for neutral ^{187}Re atoms. With the resulting $\log ft$ value of 7.87 ± 0.03 the half-life of ^{187}Re ions in any ionization state can be calculated. Thus one can correct the ^{187}Re-^{187}Os galactic chronometer calibration, by taking account of the β^- decay enhancement in stellar interiors, which will lead to a more accurate estimate of the galactic age. [S0031-9007(96)02008-X]

PACS numbers: 23.40.Hc, 27.70.+q, 29.20.Dh, 98.35.Ac

The solar abundances of most elements heavier than iron are the result of prior generations of stellar nucleosynthesis via the slow (s-) or rapid (r-) neutron-capture process. Given the effective nucleosynthesis rate, and using the abundances of long-lived radioisotopes, one can estimate a lower bound for the age of our galaxy. Compared with other chronometers like ^{232}Th and ^{238}U, the ^{187}Re-^{187}Os pair, introduced by Clayton [1], has the advantage that the long-lived nuclide ^{187}Re is produced in the r process, but not its daughter nuclide ^{187}Os. However, one uncertainty in its calibration has been pointed out by Takahashi *et al.* [2,3]: ^{187}Re may become highly ionized in the hot plasma of a star, and bound-state β^- decay may decrease the half-life from 42.3 ± 1.3 Gyr [4] by more than 9 orders of magnitude.

Bound-state β^- decay (β_b decay) is a particular weak interaction decay mode in which the decay electron remains in a bound atomic state rather than being emitted into the continuum. It is a two-body decay (the time-mirrored orbital electron capture process), in which the antineutrino carries the total decay energy (Q value) and the atomic charge state does not change. In neutral atoms, only weakly bound states are available for the decay electron and, because of their low density at the nucleus, β_b decay is only a marginal decay branch. The situation changes, when going from terrestrial conditions to a high temperature regime in stellar interiors where atoms are highly ionized and β_b decay into deeply bound orbits becomes possible. Experimentally, β_b decay was observed for the first time [5] in the case of ^{163}Dy. This nucleus is stable as a neutral atom ($Q_\beta = -2.565$ keV [6]), but, when fully ionized, it decays to ^{163}Ho ($Q_{\beta b}^K = +50.3$ keV) with a half-life [5] of 47 d.

The possible decay modes of neutral and of fully ionized ^{187}Re are illustrated in Fig. 1. For *neutral* ^{187}Re^{0+}, only the unique, first forbidden transition to the ^{187}Os ground state is energetically possible. The small matrix element and the small Q value [6] of $Q_\beta = 2.663(19)$ keV lead to the long half-life [4] of 42 Gyr. Because the inner orbits are occupied, β_b decay contributes less than 1% [7]. For *fully ionized* ^{187}Re^{75+}, β decay to continuum states is forbidden

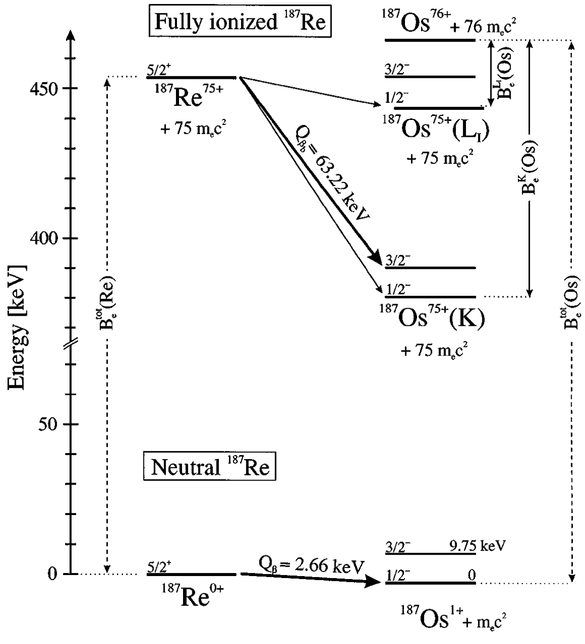

FIG. 1. Decay schemes for neutral (bottom) and fully ionized (top) ^{187}Re with the energetically allowed β transitions indicated by arrows. For *neutral* ^{187}Re^{0+}, only the unique, first forbidden transition to the ^{187}Os ground state is energetically possible. The small matrix element and the small Q value lead to the long half-life of 42 Gyr. For *fully ionized* ^{187}Re^{75+}, β_c decay (with the β particle in the continuum) is forbidden. Bare ^{187}Re^{75+} is, however, unstable against β_b decay with the electron bound in the K shell, and the dominant branch feeds the first excited state at 9.75 keV. The estimated half-life for bare ^{187}Re^{75+} is 14 yr [11].

because the electronic cloud in osmium is stronger bound by $\Delta B_e^{\mathrm{tot}} = B_e^{\mathrm{tot}}(\mathrm{Os})$-$B_e^{\mathrm{tot}}(\mathrm{Re}) = 15.31$ keV [8] than in rhenium. Instead, bare $^{187}\mathrm{Os}^{76+}$ can decay by capturing an electron from the continuum, as pointed out by Arnould [9]. Bare $^{187}\mathrm{Re}^{75+}$ is, however, unstable against β_b decay with the electron bound in the K shell and with a large Q value of $Q_{\beta b}^{K} = +72.97$ keV [10]. It was realized by Takahashi, Yokoi, and Arnould [3] that in this situation also the first excited state at 9.75 keV can be fed in a nonunique first forbidden transition with a substantially larger matrix element. The estimated half-life [11] for bare $^{187}\mathrm{Re}^{75+}$ of $T_{1/2} = 14$ yr is a billion times shorter than that for neutral $^{187}\mathrm{Re}$. An experimental confirmation of this large change in the decay probability would base the calibration of the $^{187}\mathrm{Re}$-$^{187}\mathrm{Os}$ clock on safer ground. Therefore we tried to measure the half-life of bare $^{187}\mathrm{Re}^{75+}$ nuclei in a heavy-ion storage ring by a procedure similar to that used in the first observation of β_b decay [5]. The challenge in the $^{187}\mathrm{Re}$ experiment was that the expected decay rate was about 100 times smaller than for $^{163}\mathrm{Dy}$.

$^{187}\mathrm{Re}$ ions were injected into the heavy-ion synchrotron (SIS) with a charge state $q = 50^+$, extracted at a final energy of $347A$ MeV, fully ionized to $q = 75^+$ by passing through a 100 mg/cm^2 copper foil, and finally injected into the experimental storage ring (ESR). The circulating ions were cooled by electron cooling to a small momentum spread ($\sim 10^{-5}$), diameter ($\sim$5 mm) and emittance ($\sim 0.1\pi$ mm mrad), and accumulated to coasting beams of 1–2 mA corresponding to numbers of up to 10^8 fully ionized $^{187}\mathrm{Re}^{75+}$ ions. With a residual gas pressure of 10^{-11} mbar in the ring the beam was stored with an effective half-life of 4.5 h. The losses during storage were due to collisions with the atoms of the residual gas (mainly H_2 molecules) and to changes of the charge state in the electron cooler. These loss rates have been determined not only for bare $^{187}\mathrm{Re}^{75+}$, but also for H-like $^{187}\mathrm{Re}^{74+}$ in separate experiments. With small, well-known scale factors they can also be applied to correct for the losses of H-like $^{187}\mathrm{Os}^{75+}$ ions, the daughter nuclei of $^{187}\mathrm{Re}^{75+}$ β_b decay.

With about 10^8 stored $^{187}\mathrm{Re}^{75+}$ ions, several hundred $^{187}\mathrm{Os}^{75+}$ ions were produced by β_b decay of $^{187}\mathrm{Re}^{75+}$ during storage times of up to 5 h. These $^{187}\mathrm{Os}$ ions were circulating with nearly the same revolution frequency (within 0.4 ppm) as the main beam. Their number, $N_{\mathrm{Os}}(t_s)$, grows in proportion to the storage time t_s, provided that t_s is small with respect to the half-life. For that case $N_{\mathrm{Os}}(t_s)$ is given in a good approximation by the relation [5]

$$N_{\mathrm{Os}}(t_s) = (\lambda_{\beta b}/\gamma) N_{\mathrm{Re}}(t_s) \times t_s \left(1 - \tfrac{1}{2}(\lambda_{\mathrm{Os}}^{cc} - \lambda_{\mathrm{Re}}^{cc}) t_s + \cdots\right),$$

where $N_{\mathrm{Re}}(t_s)$ denotes the number of $^{187}\mathrm{Re}^{75+}$ ions at time t_s, $\lambda_{\beta b}$ the β_b decay probability in the $^{187}\mathrm{Re}^{75+}$ rest frame, and $\gamma = E/mc^2$ the relativistic factor which takes care of the transformation from the laboratory system into the rest frame of the rapid ions. $\lambda_{\mathrm{Re}}^{cc}$ and $\lambda_{\mathrm{Os}}^{cc}$ are the loss rates of $^{187}\mathrm{Re}^{75+}$ and $^{187}\mathrm{Os}^{75+}$, respectively, due to charge changing processes and obtained as described by Jung *et al.* [5]. γ is deduced either from the revolution frequency of the ions and the circumference of the ring (about 108 m), or equivalently from the cooler electron velocity, determined by its acceleration voltage. It has been determined as $\gamma = 1.373(2)$. In order to separate the $^{187}\mathrm{Os}^{75+}$ ions from the $^{187}\mathrm{Re}^{75+}$ mother nuclei, the bound decay electron was stripped by turning on a gas jet target, which crossed the beam and produced $^{187}\mathrm{Os}^{76+}$ ions. Two methods were used to determine the number of $^{187}\mathrm{Os}^{76+}$ ions originating from the $^{187}\mathrm{Re}^{75+}$ decay.

In the first one, the Schottky-noise frequency analysis, we measured the number of circulating ions as a function of the revolution frequency [12], which allows us to identify uniquely the $^{187}\mathrm{Os}^{76+}$ daughter nuclei after the stripping process. The Schottky-noise frequency analysis is based on the fact that each circulating ion induces a signal whenever it passes a pair of capacitive pickup plates. These periodic signals reveal, when Fourier-transformed, the corresponding revolution frequency (or harmonics) of each species of stored ions. This frequency is a unique function of the mass/charge ratio since for all ions the velocity equals that of the cooler electrons. Figure 2 shows such a spectrum after storing $^{187}\mathrm{Re}$ for 1.8 h and after stripping the $^{187}\mathrm{Os}$ ions by the gas jet (turned on for 200 sec, density 3×10^{12} atoms/cm^2 of argon). For the actual measurement the primary $^{187}\mathrm{Re}^{75+}$ beam was removed from the ring, because its large intensity broadens the width of the lines from other ion

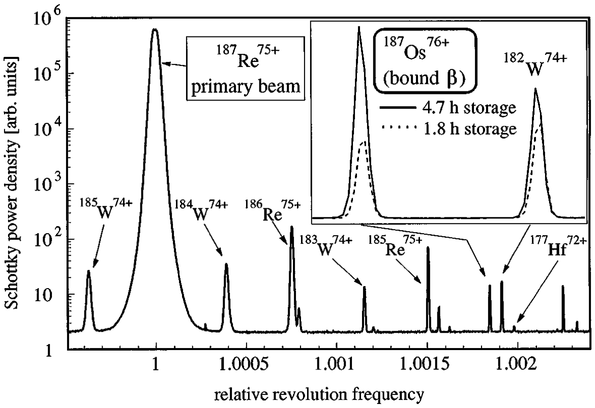

FIG. 2. Frequency spectrum of the Schottky noise after a storage time of 1.8 h and after the reaction of the coasting beam with the gas-jet target. The revolution frequency is to a good approximation a linear function of the charge/mass ratio, since all ions have the same velocity as imposed by the cooling electrons. Besides a number of nuclides produced by nuclear reactions, the β_b decay daughter $^{187}\mathrm{Os}$ can be seen. The inset demonstrates that the intensity of the $^{187}\mathrm{Os}^{76+}$ line relative to that from the nuclear reaction product $^{182}\mathrm{W}^{74+}$ increases significantly if storage time is increased from 1.8 h (dashed line) to 4.7 h (full drawn).

species by intrabeam scattering. This allows a Schottky mass spectrometry with a resolving power $M/\Delta M \approx 10^6$ [13] permitting the unambiguous identification of all nuclei of known masses circulating in the ring. All lines observed in this spectrum can be assigned to nuclei produced by reactions of ^{187}Re with nuclei in the gas jet (mainly loss of a few nucleons) except for the β_b decay daughter ^{187}Os$^{76^+}$. Only this line grows linearly with the storage time, as demonstrated in the inset of Fig. 2, proving its origin from β_b decay of ^{187}Re$^{75^+}$. A small ^{187}Os contribution originating from nuclear charge exchange reactions in ^{187}Re collisions with argon atoms of the gas jet could be determined from the intensity for zero storage time. A cross section of 0.6 ± 0.2 mb was determined, in reasonable agreement with a value of 1.3 ± 0.3 mb estimated with the intranuclear-cascade code ISApace [14]. The absolute number of ^{187}Os$^{75^+}$ ions N_{Os}, produced by the ^{187}Re β_b decay, was determined from the area of the Schottky-noise signal of fully ionized ^{187}Os$^{76^+}$ and corrected for the electron stripping efficiency of the gas jet. The latter was determined with H-like ^{187}Re$^{74^+}$ ions and scaled for $\Delta Z = 1$. The area of the Schottky lines had to be calibrated in terms of the absolute particle numbers. At high particle numbers this calibration was obtained from the relation between beam current and line area down to about 10^5 particles in the ring, corresponding to the smallest measurable current of about 2 μA. But also with very small particle numbers the calibration was possible, because the sensitivity of the detection was sufficient for observing even one single ion.

In an independent experiment we measured the position of ions that had interacted with the gas jet target and were deflected by the following dipole magnet stronger than the coasting beam. The dispersion of the magnet displaced ^{187}Os$^{76^+}$ ions from ^{187}Re$^{75^+}$ ions by 75 mm at the location of our detector. A gas microstrip counter measured the deflection position with a resolution of 0.4 mm. It was operated with 1 bar of an argon/isobutane (70:30) mixture. In front of this counter and in the same gas volume the energy loss was measured by an ionization chamber. In spite of the small energy loss of 60 MeV we achieved a resolution with respect to the nuclear charge of $\Delta Z = 1.5$. By a condition on the pulse height in the ionization chamber other elements than osmium could be suppressed in the position spectra. Figure 3 shows such a spectrum after a storage time of 4 h. The narrow peak in the middle of the spectrum is due to ^{187}Os$^{76^+}$ nuclei from the β_b decay of ^{187}Re$^{75^+}$. The background due to elastic scattering and nuclear reaction products, which have a large momentum spread and therefore a larger width in the position distribution, was measured in separate runs, in which we did not wait for the decay of ^{187}Re nuclei. The results of 10 such measurements for $\lambda_{\beta b}$ are shown as an inset in Fig. 3.

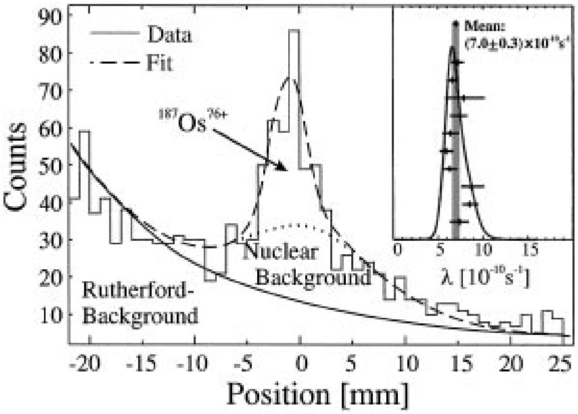

FIG. 3. Position spectrum of ions deflected by the first dipole magnet behind the gas jet target. The narrow peak in the middle of the spectrum is due to ^{187}Os$^{76^+}$ nuclei from the β_b decay of ^{187}Re$^{75^+}$. The background from elastic scattering (full drawn) and from nuclear reactions (dotted) has been determined in separate runs. The inset shows the results of ten individual measurements for the decay constant in form of an ideogram, as prescribed in the Review of Particle Properties [15]. The full drawn curve, the sum of Gaussians for each measurement, serves mainly to judge the consistency of the data.

With either method a dozen measurements were performed with storage times t_s, ranging from $t_s \approx 0$ (determination of background from nuclear reactions) to $t_s \approx 5$ h. After calibrating the areas of the Schottky signals and of the line in the particle detector in absolute particle numbers and after determining the number of primary ^{187}Re$^{75^+}$ ions (with a beam current transformer), taking into account the various losses, we obtain for the β_b decay probability $\lambda_{\beta b}$ of bare ^{187}Re in the ion rest frame the two independent results $\lambda_{\beta b} = (6.29 \pm 0.19 \pm 0.40) \times 10^{-10}$ s^{-1} from the Schottky-noise analysis and $\lambda_{\beta b} = (7.05 \pm 0.28 \pm 0.34) \times 10^{-10}$ s^{-1} from the position spectra, where first the statistical errors are given and then the estimated systematical errors. Adding these two errors algebraically in each case and then taking the average of both results, we get $\langle \lambda_{\beta b} \rangle = (6.7 \pm 0.4) \times 10^{-10}$ s^{-1} and $T_{1/2} = (32.9 \pm 2.0)$ yr. The measured $\lambda_{\beta b}$ is practically equal to the β_b decay probability into the K shell of ^{187}Os, because the decay into the L shell is about 4 orders of magnitude less probable. From the measured $T_{1/2}$ we deduce $\log ft = 7.87 \pm 0.03$. Note also, that the decay of bare ^{187}Re is dominated by the nonunique transition to the first excited state of ^{187}Os, since the decay to the ground state has a much smaller matrix element ($\log ft = 11.0$, from the decay of neutral ^{187}Re). According to Ref. [2] Re ions in a stellar plasma can have average numbers of bound electrons between 1 and more than 20, depending on temperature and density. With this measured ft value the decay rate of ^{187}Re in any charge state, and hence at any temperature, can now be calculated.

Our measurement is very close to $\log ft = 7.5$, which was assumed by Yokoi *et al.* [3] in their study of the ^{187}Re-^{187}Os cosmochronometry. In that work they dealt with various problems: (1) The history of nucleosynthesis over the galactic time scale had to be described by a chemical evolution model, a common requirement for all cosmochronometers. (2) The transformation of already existing ^{187}Re and ^{187}Os into other nuclides, when embedded into the hot environment in next generation stars (commonly called "astration"). (3) The production of ^{187}Os other than from ^{187}Re β decay, namely, by neutron capture reactions in the s process. This is normally assumed to be proportional to the s process production of ^{186}Os, but modifications due to neutron capture on the excited state of ^{187}Os and modifications in the s-process path could only be estimated [16,17].

One of the major uncertainties concerned the evaluation of β-transmutation rates. Now, with the measured nuclear matrix element of the key β transition one may hope to remove that uncertainty. The ^{187}Re decay rate was overestimated by Yokoi *et al.* [3] only by a factor of about 2.3. As this factor also applies to the reverse transition, the electron capture decay of ^{187}Os from the thermally excited 9.75 keV state, the net effect of our measured value for the transition rate would be small: the galactic age (estimated in the range from 11 to 15 Gyr) would increase by less than 1 Gyr. On the other hand, the work of Yokoi *et al.* adopted a simple model of stellar evolution. In the past decade many systematic studies of stellar evolution have been made. Therefore it seems timely to improve the estimates on the astration effects. Similarly, astronomical observational data have been accumulated that have to be used as constraints in modeling the galactic chemical evolution. The other nuclear physics quantity important for Re-Os chronology—besides the β decay matrix element determined in this work—is the neutron capture cross section of excited ^{187}Os, which in the meantime can be estimated within 5% by using related experimental information [18]. The last major problem is the abundance data on Re and Os that are still uncertain by 9% and 6%, respectively [19]. In attacking all these problems, one may hope that the Re-Os pair—the most promising nuclear cosmochronometer after all—will reveal a reliable galactic age.

We sincerely thank the operators and engineers of the GSI accelerators for providing us with an excellent ^{187}Re beam over weeks. We are indebted to K. Sümmerer for providing us with the ^{187}Re/^{187}Os charge-exchange cross section calculated with the ISApace code. This work was supported by the "Sonderforschungsbereich 375-95 für Astro-Teilchenphysik der Deutschen Forschungsgemeinschaft" and the accelerator laboratory of the Munich universities.

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