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BOUND-STATE DECAY OF RHENIUM-187

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ABSTRACT

We have calculated the ratio of bound-state to continuum β-decay of the potential cosmochronometer ^{187}Re using Dirac wavefunctions in a Thomas-Fermi atomic potential, including exchange corrections. We found $\lesssim 1\%$ bound-state decay for the neutral atom. This differs from a previous calculation using hydrogenic wavefunctions (58%), and from a comparison of published laboratory and geochemical measurements (53% $\pm$ 35%).

Subject headings: abundances — nuclear reactions — nucleosynthesis — transition probabilities

I. INTRODUCTION

The half-life of the low-energy (2.64 keV) β-decay of ^{187}Re to ^{187}Os is of critical importance in the application of the Re/Os chronology (Clayton 1964; Woosley and Fowler 1979) to the determination of the duration of r-process nucleosynthesis in the Galaxy prior to the formation of the solar system. The value of the half-life is currently a matter of some controversy, namely that involving the ratio of bound-state to continuum β-decay for ^{187}Re. By this we mean the fraction of decays in which the final Os atom is neutral, rather than being positively charged, as would be the case if the β-ray were emitted into the continuum.

The most recent geochemical determination of the half-life of the natural β-decay of ^{187}Re was performed by Luck and Allègre (1983) on several iron meteorites and on the metallic phases of chondrites and yielded 45.6 $\pm$ 1.2 Gyr. This determination agrees well with the value 43 $\pm$ 5 Gyr measured in molybdenite ores by Hirt *et al.* (1963), who summarize previous less accurate measurements. It is presumed that the geochemical determination involving ratios of the daughter ^{187}Os to the parent ^{187}Re measures the combined half-life due to both continuum and bound-state decay.

Two direct measurements involving the detection of low-energy electrons have been made, and of course these measure only the half-life for continuum decay. Brodzinski and Conway (1965) reported the half-life to be 66 $\pm$ 13 Gyr, and subsequently Perrone (1971) found theoretically a ratio of bound-state to continuum decay of 58% (with considerable uncertainty), which agrees with the value 53% $\pm$ 35% derivable from the results of Hirt *et al.* and Brodzinski and Conway.

At the same time as the Brodzinski and Conway work, however, Payne (1965) measured the half-life as 47 $\pm$ 5 Gyr. When combined with the most recent geochemical determination (Luck and Allègre 1983) of 45.6 $\pm$ 1.2 Gyr, this implies a bound-state to continuum decay ratio of 3% $\pm$ 2%.

In addition to its importance in nucleosynthesis, this low-energy β decay is the best candidate for measurement of any long-term change in the fine-structure constant α (Dyson 1972). The low endpoint energy of the decay is the result of a near cancelation of the differences of Coulomb and nuclear energies between rhenium and osmium, so that a fractional change in α changes the Coulomb energy difference, which changes the endpoint energy, and so the half-life. The fractional change in half-life is $\sim$18,000 times the fractional change in α, so that an accurate knowledge of the geochemical and laboratory half-lives may be relevant here.

In what follows we report a new theoretical analysis resulting in a value of at most 1% for the ratio of bound-state to continuum decay.

II. CALCULATION

The β-transition of neutral atomic ^{187}Re to ^{187}Os has endpoint energy $E_0 = 2.64$ keV and is first-forbidden unique, $5/2^+ \rightarrow 1/2^-$. Since it is unique, only one nuclear matrix element is involved, and we need only electron wave functions and phase-space considerations to calculate the ratio of bound to continuum decay.

The vector sum of electron and (anti)neutrino angular momenta must be 2, and their parities opposite. The β-decay amplitude is proportional to each of the lepton wave functions at the nuclear surface, and these are centrifugally suppressed by the factor $(pR)^l$, where $\hbar p$ is the lepton momentum, R is the nuclear radius, and l is the orbital angular momentum. Since $pR \ll 1$, we use the normal approximation, with $l_e + l_\nu$ as small as possible consistent with the selection rules. Thus, one lepton is in a $p_{3/2}$ state, and the other in an $s_{1/2}$ state. The electron momentum is dominated by the Coulomb contribution, and is $\sim Ze^2/Rc \approx 15$ MeV/c, so that the neutrino momentum, $\sim E_0/c \approx 2.64$ keV/c, is much smaller. Thus we expect the dominant contribution to the decay amplitude is from $e(p_{3/2})$, $\bar{\nu}(s_{1/2})$, rather than $\bar{\nu}(p_{3/2})$, $e(s_{1/2})$.

We adopt natural units $\hbar = c = m_e = 1$, and write the lepton spinor in the conventional way (Rose 1961),

$$\varphi(r) = \begin{pmatrix} g_\kappa(r)\chi_{\kappa m}(\hat{r}) \\ if_\kappa(r)\chi_{-\kappa m}(\hat{r}) \end{pmatrix}, \tag{1}$$

where $\chi_{\kappa m}$ is a two-component spin-spherical harmonic. For a $p_{3/2}$ state, $\kappa = -2$, and for a $s_{1/2}$ state, $\kappa = -1$. The large and small radial wave functions g_κ and f_κ satisfy the coupled radial Dirac equations,

$$\begin{aligned} \frac{df_\kappa}{dr} &= \frac{\kappa - 1}{r} f_\kappa - [W - 1 - V(r)]g_\kappa \\ \frac{dg_\kappa}{dr} &= [W - V(r) + 1]f_\kappa - \frac{\kappa + 1}{r} g_\kappa , \end{aligned} \tag{2}$$

where $V(r)$ is zero for the neutrino and is for the electron the sum of the electrostatic potentials of the nucleus and of the atomic electrons.

The continuum decay rate is proportional to the f_1 value for the transition, given by Gove and Martin (1971),

$$f_1 = \int_1^{W_0} \frac{9W}{2pR^2} (W_0 - W)^2 |g_{-2}{}^W(R)|^2 dW , \tag{3}$$

where W_0 is the total endpoint energy $1 + E_0$, R is the nuclear radius, $p = (W^2 - 1)^{1/2}$ is the electron wavenumber, and $g_{-2}{}^W(r)$ is the large component of the electron final state spinor. This continuum spinor is normalized so that its asymptotic behavior is

$$\begin{aligned} g_\kappa{}^W &= \frac{1}{r}\left(\frac{W+1}{W}\right)^{1/2} \cos [pr + \delta(r)] \\ f_\kappa{}^W &= \frac{1}{r}\left(\frac{W-1}{W}\right)^{1/2} \sin [pr + \delta(r)] . \end{aligned} \tag{4}$$

We have omitted terms from equation (3) corresponding to other angular momenta for the electron and neutrino final states, but these can be neglected by the arguments above. We calculated that the term for $\bar{\nu}(p_{3/2})$, $e(s_{1/2})$ contributes $\sim 1\%$ of the term in equation (3).

We shall now calculate $f_1{}^B$, the corresponding rate for bound-state decay. For the bound-state Dirac spinors $g_{-2}{}^n(r)$ we adopt the usual normalization

$$\int_0^\infty r^2 dr[|g_{-2}{}^n(r)|^2 + |f_{-2}{}^n(r)|^2] = 1 . \tag{5}$$

Now suppose the atom to be within a large sphere of radius L with hard walls, so that bound and continuum states can be treated on the same footing. The continuum spinors have the form $L^{-1/2} g_{-2}{}^W(r)$ in order to be correctly normalized as a bound state. They have asymptotic form proportional to $r^{-1} \sin (pr + \delta)$, so the states are separated by $\delta p = \pi/L$, so the density of states is L/π per unit p, which is $LW/\pi p$ per unit energy. We can thus replace $\int (LW/\pi p)dW$ by a sum over bound states. From equation (3).

$$f_1 = \int_1^{W_0} \frac{LW}{\pi p} dW \frac{9\pi}{2R^2} (W_0 - W)^2 |g_{-2}{}^W(R) L^{-1/2}|^2 \tag{6}$$

$$\rightarrow f_1{}^B = \sum_n \frac{9\pi}{2R^2} (W_0 - W_n)^2 |g_{-2}{}^n(R)|^2 , \tag{7}$$

where n labels an allowed $p_{3/2}$ bound state in which the electron can be created, and W_n is its (total) binding energy. For the Re/Os system, the $6p_{3/2}$ and subsequent shells are vacant.

Equation (7) gives the $f_1{}^B$ value for an electron to be created directly into a vacant orbital. There are exchange and overlap effects however (Bahcall 1965), which Gilbert (1958) estimated (using Slater orbitals) to be about 9 times the direct contribution. These occur because the nuclear charge changes in the decay, so that there is an imperfect overlap between the Re and Os^+ atomic states. For consistency with the continuum part of the calculation, we shall treat the final state as an Os^+ ion with an extra electron, rather than a neutral Os atom. If the Re atom is initially in its ground state, filled to the Fermi level, and we use these Os^+ basis states, there is a nonzero amplitude for the Os^+ atom to have a hole below the Fermi level and an electron above, so the β-electron can be created in the hole. For each particle state, these contributions (summed over the various possible hole states) add coherently to the direct amplitude for decay into that particle state. The rates for different particle states (final states of the Os^+) then add incoherently, in the same way as in equation (7). Let the label p refer to particle (vacant) states, and h to hole (occupied) states. Writing $|0\rangle$ for the Os^+ ground state, the Os state after the β-decay is then $|f\rangle = a_p{}^+|0\rangle$. The Hamiltonian can be written as

$$H_\beta = \sum_{n=h,p} a_n{}^+ g_{-2}{}^n(R) , \tag{8}$$

and the initial Re state is approximately

$$|i\rangle = \left(1 + \sum_{ph} a_p{}^+ a_h \langle p, \mathrm{Os}^+ | h, \mathrm{Re}\rangle\right)|0\rangle . \tag{9}$$

The $f_1{}^B$ value is then

$$f_1{}^B = \sum_p \left| g_{-2}{}^p(R) - \sum_h \langle p, \mathrm{Os}^+ | h, \mathrm{Re}\rangle g_{-2}{}^h(R)\right|^2 \frac{9\pi}{2R^2} (W_0 - 1)^2 . \tag{10}$$

We have here approximated the atomic binding energies (a few eV) to be negligible in comparison to the endpoint energy (2.64 keV). In principle there is a similar exchange contribution to the continuum decay rate, but this is negligible for the following reason. The exchange continuum contribution to the rate will be less than the exchange bound-state contribution because the overlaps will be smaller. We find the direct bound and exchange bound contributions to be of the same order, and the direct continuum to be ~ 100 times larger; thus the continuum exchange is a $\sim 1\%$ correction to the continuum rate, which is negligible.

We can now approximately reproduce Perrone's result. He only considered the direct contributions, using nonrelativistic screened hydrogenic wavefunctions with $Z = 61$. Then the form of the wavefunctions at the origin is known explicitly, and we can write equation (7) as

$$f_1^B = \sum_{6}^{\infty} 2\pi(Z\alpha)^5 n^{-3}(W_0 - 1)^2 \,, \tag{11}$$

so $\log f_1^B = -7.31$. The "screened" $\log f_1$ value for $Z = 61$ and $E_0 = 2.64$ keV is -7.29, from Gove and Martin, so the bound-state decay fraction is 49%, compared to Perrone's 58%. We shall now show that a better description of the atomic potential leads to a much smaller bound-state decay fraction.

For the potential $V(r)$ in equation (2), we used the Lenz-Jensen potential (Gombas 1949), which is an analytic approximation to the Thomas-Fermi model,

$$V_{LJ}(r, Z) = -C(r, Z)e^{-x}(1 + x + b_2 x^2 + b_3 x^3 + b_4 x^4) \,,$$
where
$$b_2 = 0.3344 \,, \quad b_3 = 0.0484 \,, \quad \text{and} \quad b_4 = 2.647 \times 10^{-3} \,. \tag{12}$$

The variable $x = (10.9\alpha r)^{1/2} Z^{1/6}$, where Z is the nuclear charge, and $C(r, Z)$ is the nuclear Coulomb potential, taken to be that of a uniformly charged sphere of radius R,

$$C(r, Z) = \begin{cases} (Z\alpha/2R)[3 - (r/R)^2] & r < R \\ Z(\alpha/r) & r \geq R \end{cases} . \tag{13}$$

We took the nuclear radius to be 6.75 fm $= 1.18A^{1/3}$ fm.

The wave function of the bound state at the nuclear surface is sensitive to the binding energy of the state, and since this is not accurately reproduced by the Lenz-Jensen potential for the unoccupied valence states in which the electron is created, we added an adjustable Coulomb tail to the Lenz-Jensen potential:

$$V(r) = V_{LJ}(r, Z = 75 - Z_\infty) - C(r, Z_\infty) \,. \tag{14}$$

We can thus get the correct binding energy for one of the valence electrons and presume that the energies of the unoccupied states are then correct. We take equation (14) as the form of the Os^+ potential, so the Re potential is $V(r) - C(r, 1)$.

To solve the coupled Dirac equations, we used a fifth-order Runge-Kutta procedure in exponential coordinate $r = e^\rho$, with lower bound corresponding to 0.1 R. For the continuum states, we integrated out until the average $W/(W + 1)r^2 |g_{-2}^W|^2 + W/(W - 1)r^2 |f_{-2}^W|^2$ was constant, then computed the normalization from this average. We computed the $\log f_1$ value by Simpson's rule with 50 points for the Lenz-Jensen potential with three values of Z_∞ (see below), and for a pure Coulomb potential, and it was -6.85 for all four cases, which agrees with the tables of Gove and Martin.

For the bound states, we used a binary search in energy as follows. Beginning with a pair of energy values bracketing the required bound-state energy, and integrating out well beyond the classically allowed region, one wavefunction behaved like $e^{\rho r}$ and the other like $-e^{\rho r}$ at large distances. The energy interval is then halved at each step on the basis of the behavior of the wavefunction at the energy in the middle of the bracket. For the overlaps and normalizations of the Re and Os^+ states, we used essentially the same procedure, but with two simultaneous binary searches while computing the overlap and normalization integrals by adding extra equations to the differential equation set.

Table 1 shows the calculation according to equation (10) for $Z_\infty = 2$. The first column is the principal quantum number in the $p_{3/2}$ shell in the Re atom, labeled h, and the second is the binding energy in eV of an electron in the Re atom for that shell. The third column is the direct contribution to the $\log f_1^B$ value from that shell if it were vacant. The last three columns are the values of the

TABLE 1

THE CALCULATION OF THE LOG f_1^B VALUE FOR $Z_\infty = 2$

h	Re $p_{3/2}$ ENERGY (eV)	LOG f_1^B	OVERLAPS $p = 6$	$p = 7$	$p = 8$
2	10390	−5.78	0.00023	0.00013	0.00004
3	2220	−6.35	0.00095	0.00055	0.00016
4	360	−6.98	0.00511	0.00293	0.00089
5	18.4	−7.86	0.0767	0.0420	0.0122
6	2.60	−9.03	1	…	…
7	1.24	−9.84	…	1	…
8	0.25	−10.51	…	…	1
$\log f_1^B$ (direct)		−8.95			
$\log f_{1h}^B$			−9.45	−10.50	−10.87
$\log f_1^B$ (direct + exchange)					−9.40

TABLE 2

RESULTS FOR VARIOUS Z_∞

Z_∞	2	2.4	2.75	Exp.
Re $5p_{3/2}$				
energy (eV)	18.4	26.9	34.6	34.6 ± 0.6
Re $5d_{5/2}$				
energy (eV)	1.6	3.4	6.3	3.5 ± 0.5
Direct:				
$\log f_1^B$	−8.95	−8.72	−8.62	...
Bound decay	0.78%	1.33%	1.67%	...
Direct + Exchange:				
$\log f_1^B$	−9.40	−9.12	−9.00	...
Bound decay	0.28%	0.54%	0.70%	...

NOTE.—The continuum $\log f_1$ value is -6.85, and the bound decay fractions are percentages. The experimental energy levels in the last column are from Bearden and Burr.

overlaps of the $n = 6$, 7, and 8 states in Os^+, labeled p, with the Re state of the first column. Below the third column is the direct log f_1^B value, obtained by summing the f_1^B values for $h = 6$, 7, and 8. Below the last three columns is the (direct + exchange) $\log f_1^B$ value for each p state, with the total below. Notice that the exchange terms destructively interfere with the direct, making a lower f_1^B value. The value $Z_\infty = 2$ corresponds to the potential felt by an electron far removed from the Os^+ ion. This value of Z_∞ gives the binding energy of the $5p$ state of Re as 18.6 eV, much smaller than the measured value of ~ 34.6 eV (Bearden and Burr 1967). We found that $Z_\infty = 2.75$ reproduces this energy. Alternatively, we tried to get the $5d_{5/2}$ state correct, at ~ 3.5 eV (Bearden and Burr 1967), resulting in a value $Z_\infty = 2.4$. Table 2 shows the results for the three values of Z_∞, together with the energy levels in the Re atom corresponding to these potentials. When trying to reproduce a more tightly bound state, Z_∞ is larger; this is because the screening of the nuclear charge is less. Since the bound-state decay creates an electron in a vacant orbital, it is these that we wish to describe accurately, and consequently the "asymptotic" potential with $Z_\infty = 2$ is probably the most accurate.

III. CONCLUSIONS

We have shown that the bound-state decay fraction of ^{187}Re is less than 1% by using more accurate electron orbitals instead of the hydrogenic orbitals used in previous work (Perrone 1971; Gilbert 1958). We have neglected the effects of the Coulomb interaction of the ejected β-ray with the atomic electron (Williams and Koonin 1983), which may cause shake-up or shake-off in the atom. We feel that these effects are unlikely to significantly change the possible bound-state orbitals available to the β-electron, and thus not to change our result much.

Our calculation deals with neutral atomic Re; there is a considerable uncertainty in the Re/Os chronology because the half-life of ^{187}Re decreases dramatically when it is ionized under astrophysical conditions in the interior of stars. This has been extensively discussed by Takahashi and Yokoi (1983).

There is additional uncertainty in the Re/Os chronology arising in part from the fact that the amount of radiogenic ^{187}Os must be corrected for that produced independently by the s-process. The value of σN_s is constant along an s-process chain, where σ is the neutron capture cross section for a particular isotope and N_s the abundance of the isotope. The cross section for the ground state of ^{187}Os has been measured in the laboratory; unfortunately the excited state of ^{187}Os at 9.75 keV is also populated in the s-process (at $kT \approx 30$ keV), and the neutron capture on this excited state cannot be directly determined in the laboratory. Indirect measurements are underway in a number of laboratories, and the final outcome of the Re/Os chronology of the Galaxy awaits these results.

We have shown that the bound state decay of ^{187}Re is less than 1% of the total and thus that the geochemical and most precise laboratory measurements are consistent. Since the former are considerably more precise than the latter, we recommend the value 45.6 ± 1.2 Gyr for the half-life of neutral ^{187}Re.

Note added in manuscript.—It has come to our attention that Borg and Lindner at Lawrence Livermore National Laboratory are making a new direct measurement of the ^{187}Re half-life.

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