## **Neutrinos: Ghosts of creation**

## J R de Laeter

Department of Applied Physics, Curtin University of Technology, Perth WA 6001

#### Abstract

The magnitude of the mass of an electron neutrino is still uncertain, yet this parameter is of vital importance to our understanding of cosmology, particle physics and neutrino oscillations. The process of double beta decay offers a means of testing grand unification theories as it may yield limits for the electron neutrino mass and indicate the degree of lepton conservation in elementary particle reactions. A number of measurements of the half-life of double beta decay of <sup>130,128</sup>Te/<sup>130,128</sup>Xe and <sup>82</sup>Se/<sup>82</sup>Kr have been obtained, but the values are almost certainly inaccurate because of leakage of the gaseous daughters over geological time. This may be overcome by measuring the double beta decay half-lives of nuclides which have solid daughter products. Improvements in solid source mass spectrometry now allow such measurements to be successfully attempted. The results of one such experiment will be described and other double beta decay systems which may yield experimental results suggested.

## Introduction

On December 4<sup>th</sup> 1930, the Austrian physicist Wolfgang Pauli suggested that a particle might be responsible for carrying off the varying amounts of energy that seemed to be missing in beta decay. However, over two decades were to elapse before neutrinos were finally discovered by Reines & Cowan (1956). It has since been shown that there are different types of neutrinos - electron neutrinos which are emitted during beta decay, muon neutrinos which are associated with charged pion decay, and tau neutrinos.

The only way that neutrinos can interact with normal matter is through an extremely feeble force known as the weak interaction. Thus, in the "standard" model of particle physics, neutrinos are mass-less particles that do not disintegrate. There is some experimental evidence (albeit controversial) that neutrinos can oscillate from one form to another (Athanassopoulos *et al.* 1995). This can only occur if these neutrinos possess a mass. In 1987, supernova SN 1987A exploded in the Large Magellanic Cloud. From the spread in the arrival times of the burst of neutrinos emitted by SN 1987A, it could be calculated that the mass of the electron neutrino is less than 25eV.

One of the most successful developments of modern science has been the rapid evolution of nuclear astrophysics to what is now a mature scientific study. A study of nuclear astrophysics has enabled us to unravel many of the secrets of nucleosynthesis – those nuclear constraints which determine the abundances of the chemical elements. The isotope abundances of the elements, which we measure so precisely by mass spectrometry today, have been fashioned in the stars and interstellar medium over billions of years. Some are vestiges of the Big Bang itself. The three basic process involved in synthesising the heavy elements (Z>31) are the slow (s) and rapid (r) neutron capture processes, and the proton (p) process, a reaction that synthesises the neutron-deficient isotopes. The s-process synthesises nuclides along the valley of nuclear stability during the red giant phase of a star's evolution, whilst the r and p processes occur on a much shorter time scale in cataclysmic, non-equilibrium environments, such as supernova. De Laeter (1990) has described these nucleosynthetic processes in greater detail.

Figure 1 shows a section of the Chart of the Nuclides in the vicinity of tin. The r-process nuclides mimic fission product decay chains and produce neutron-rich isotopes, in some cases far from the valley of stability. From the nuclear stability of these r-process even-even nuclides, it can be shown that they cannot be absolutely stable, yet the fact remains that they have sustained their existence in measurable quantities over geological time. There is therefore a need for a nuclear decay mechanism to exist that is sufficiently slow to enable these nuclides to survive in significant quantities over a period of many billion years.

Double beta decay is just such a mechanism in that its half-life is in excess of  $10^{17}$  years (Mayer 1935). The nuclear systematics of double beta decay is applicable to even-even nuclides on the neutron-rich side of the valley of nuclear stability, thus enabling survival of these nuclides over geological time scales.

## **Double beta decay**

Double beta decay is the rarest nuclear phenomenon in Nature. In double beta decay, a nucleus undergoes a transmutation from one element to another such that two electrons are emitted rather than the single electron emitted, in the more commonly observed single beta decay. All of the parent and daughter isotopes involved in double beta decay are even-even nuclei. The pairing force acting between like nucleons is responsible for the

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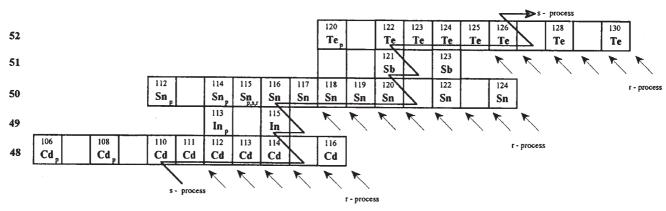


Figure 1. A section of the Chart of the Nuclides in the vicinity of tin. The arrows represent nuclides produced by the r-process, which indicates the various modes of nucleosynthesis.

increase in the binding energy of these nuclei relative to the odd-odd isotopes, thus preventing a single beta decay.

The long standing question in double beta decay is whether the electron neutrino should be described by a Dirac or by a Majorana field (Primakoff & Rosen 1981). In the Dirac formalism, the neutrino has a distinct antiparticle such that:

$$(A, Z) \rightarrow (A, Z + 2) + 2e^{-} + 2\overline{\nu}_{a}$$

However, if the neutrino is a Majorana particle then the neutrino and anti-neutrino are indistinguishable, so that double beta decay can occur with the net emission of no neutrinos such that;

$$(A, Z) \rightarrow (A, Z + 2) + 2e^{-2}$$

Grand Unified Theories of particle physics allow the neutrino to have a finite mass, thus allowing the possibility of neutrino-less double beta decay. However, double beta decay systems have such long half lives  $(10^{17} - 10^{24}$  years; Doi *et al.* 1985), that enormous experimental constraints are imposed on the detection of double beta decay daughter products.

There are two approaches to measuring the half-life of a double beta decay reaction. In the direct detection method, coincidence counting is used to measure the radioactivity at the energy value predicted from precise atomic mass measurements (Dyck et al. 1990). The disadvantage of the radioactive counting method is that intrinsically faster radioactivities, such as those associated with uranium or thorium decay chains, produce significant backgrounds which may mask the beta decay product. Instrumental experiments are designed to detect the electrons as they are emitted and are thus capable of not only measuring the lifetime of the double beta decay, but also the electron energy spectrum. Doi et al. (1988) have summarised a number of experimental results for the half-life of double beta decay on <sup>76</sup>Ge, <sup>82</sup>Se, <sup>100</sup>Mo, <sup>136</sup>Xe and <sup>150</sup>Nd conducted by a number of research groups (Table 1). The half lives range from  $>6 \times 10^{18}$  years to  $>1.4 \text{ x } 10^{21} \text{ years.}$ 

One of these direct detection experiments was conducted by the University of California, Santa Barbara and Lawrence Berkeley Livermore, 600m underground with eight 0.9kg Ge detectors. The detectors are inside a cavity formed by 10 blocks of 15 cm thick sodium iodide crystals providing an active volume with a 30 keV threshold. The sodium iodide crystals are in turn inside

#### Table 1

The data of the half-lives of the neutrinoless double-beta decay with Majorana emission for various nuclei. (Doi 1988 and primary references therein).

	Experimental group	Half-life (yr)
<sup>76</sup> Ge	Osaka	>2 1020
	Moscow (ITEP)	$>2 \ 10^{20}$
	Batelle-South Carolina	$6 (\pm 1) 10^{20}$
	Caltech-SIN-Neuchatel	>12 10 <sup>20</sup>
	Santa Barbara-LBL	>14 10 <sup>20</sup>
<sup>82</sup> Se	Irvine	>4.4 10 <sup>20</sup>
<sup>100</sup> Mo	Osaka	>6 1018
	Irvine	>7.5 1018
<sup>136</sup> Xe	Milano	>1.6 1019
	Moscow (INR)	>1.0 10 <sup>20</sup>
$^{150}Nd$	Moscow (INR)	>7.0 1019

a pure Pb shield of 20 cm thickness. The detectors are kept at liquid-nitrogen temperatures under vacuum. The detectors are tuned to search for electrons emitted from the <sup>76</sup>Ge  $\rightarrow$  <sup>76</sup>Se double beta decay with a 2.041 MeV summed electron energy, which is the total kinetic energy available for this particular reaction.

The second approach to measuring the half-life of a double beta decay reaction is based on the cumulative effects of double beta decay over geological time. This geochemical method depends on the mass spectrometric determination of the stable double beta decay product that has accumulated in geologically old minerals. The geochemical method does not directly determine the decay mode as is possible in the direct determination method. It indicates only the total decay probability of two neutrino decay  $\lambda_{2\nu}$  and no neutrino decay  $\lambda_{0\nu}$ ,  $\lambda_{2\nu} + \lambda_{0\nu}$ . However, for double beta decay accompanied by the emission of two neutrinos, the theoretically predicted decay probability  $\lambda_{2\nu}$  is much smaller than that predicted in neutrinoless double beta decay  $\lambda_{0\nu}$  by perhaps six orders of magnitude (Doi *et al.* 1985), although the actual value depends on the mass of the neutrino.

The geochemical method provides an upper limit to the double beta decay half-life because the gas retention age invariably post-dates the time of mineralisation; the direct detection method yields lower limits because of the possibility of background events. The direct detection

method has the potential advantage of being able to distinguish between the two double beta decay modes. In the case of the two neutrino double beta decay mode, the energy deposited in the detectors will show a continuous energy distribution up to the total transition energy, in the same manner as single beta decay. However, if neutrino-less double beta decay occurs, the sum of the energies of the two emitted electrons will be the total transition energy, and the energy spectrum will be a single narrow peak. The energy spectrum therefore provides a clear, unambiguous signature of the nature of double beta decay. The high resolution, double focusing mass spectrometer at the University of Manitoba has been used to determine double beta decay energy values for <sup>128</sup>Te and <sup>130</sup>Te of 867.2  $\pm$  1.0 and 2528.8  $\pm$  1.3 keV respectively (Dyck et al. 1990). The total kinetic energy available for any double beta decay system can be obtained from the atomic mass table of Wapstra & Audi (1985).

# Geochemical half-life determinations of double beta decay

The geochemical method of measuring double beta decay depends on the accumulation of decay products from this rare nuclear process in a natural mineral of the parent element. The method is most likely to succeed if little of the daughter element was incorporated when the mineral of the parent element formed. The likelihood of this separation occurring in nature is enhanced if the geochemical nature of the double beta decay product is radically different from that of the parent element. Success also depends on the mineral being geologically old and having survived intact over a long period of time so that detectable quantities of the daughter have accumulated in the mineral. Hard, high temperature minerals have a favoured chance of surviving temperature/pressure variations to which the minerals may have been exposed during their geological history.

Geochemical measurements of double beta decay have, until recently, been confined to the measurement of gaseous daughter products (by gas source mass spectrometry), which have accumulated in old tellurium and selenium minerals from the following reactions;

19037

<sup>130</sup> Ге	$\rightarrow$	ββ	$\rightarrow$	<sup>130</sup> Xe	+	$2\overline{\nu}_{e}$
<sup>128</sup> Te	$\rightarrow$	ββ	$\rightarrow$	<sup>128</sup> Xe	+	$2\overline{\nu}_{ m e}$
<sup>82</sup> Se	$\rightarrow$	ββ	$\rightarrow$	<sup>82</sup> Kr	+	$2\overline{\nu}_{e}$

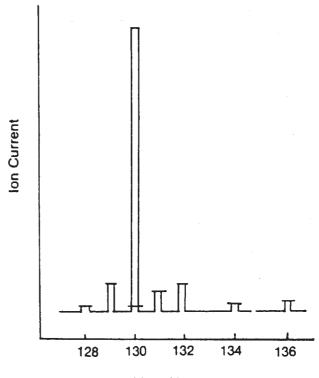
The double beta decay of <sup>130</sup>Te to <sup>130</sup>Xe is a favourable experimental situation because <sup>130</sup>Te is the most abundant nuclide of Te, old telluride minerals of high concentration are available, and the mass spectrometry of Xe can be carried out with high sensitivity. Inghram & Reynolds (1949) first estimated the half-life of <sup>130</sup>Te for double beta decay to be 1.4 x 10<sup>21</sup>y. Since that time, many <sup>130</sup>Te decay experiments have been carried out and a partial list of the results are given in Table 2. The current best estimate of the half-life of <sup>130</sup>Te is 8 10<sup>20</sup>y (Manuel 1991). Figure 2 (taken from Kirsten *et al.* 1968) shows the dominance of <sup>130</sup>Xe in the mass spectrum of Xe extracted from an old telluride ore sample. Similar mass spectrometric determinations have been made for the double beta decay half-life of <sup>128</sup>Te and <sup>82</sup>Se. Manuel

(1991) gives best estimates of these half-lives as  $2 \ 10^{24}$ y and  $1 \ 10^{20}$ y respectively. The results from these isotopic systems support the lepton-conserving, two neutrino mode of decay.

Table 2

Values reported for the half-life of 130Te

Reference	Half-life 1.4 10 <sup>21</sup>		
Inghram & Reynolds (1949)			
Takaoka & Ogata, (1966)	8.2 1020		
Srinivasan et al. (1972)	2.5 1021		
Hennecke et al. (1975)	9.7 1020		
Kirsten (1983)	2.6 1021		
Richardson et al. (1986)	<1 10 <sup>21</sup>		
Kirsten et al. (1986)	1.6 1021		
Manuel (1986)	7 1020		
Chiou & Manuel (1988)	7 1020		
Current best estimate	8 10 <sup>20</sup>		



Mass Number A

**Figure 2.** Isotopic composition of xenon extracted from tellurium ore (Kirsten *et al.* 1968). The horizontal lines indicate the maximum contribution of atmospheric xenon.

The major analytical shortcoming of the Te/Xe and Se/Kr decay systems is that their daughters are gaseous and therefore may have suffered some leakage from the ore sample over the long time period since the minerals were formed. Minerals of tellurium and selenium are particularly susceptible to gas loss because they are soft, low temperature minerals that may have recrystallised and lost radiogenic Xe or Kr during any deformation event or high temperature phase subsequent to mineralisation. Richardson *et al.* (1986) have shown that gas retention ages are significantly less than mineralisation ages. Lin *et al.* (1986) discuss the problems involved with gas retention in the mineral kitkaite.

Although there may well be uncertainties in the absolute values of <sup>128</sup>Te, <sup>130</sup>Te and <sup>82</sup>Se, there are compelling reasons why the ratio of the half-lives of <sup>128</sup>Te, <sup>130</sup>Te for double beta decay should be determined. The great advantage of the Te/Xe double beta decay system is that both the daughter products are noble gases and the ratio <sup>128</sup>Xe/<sup>130</sup>Xe, which is directly proportional to the ratio of the half lives of <sup>128</sup>Te/<sup>130</sup>Te, can be determined with relatively high precision by noble gas mass spectrometry, thus eliminating many sources of systematic error.

Pontecorvo (1968) has shown that the ratio of the decay rates of pairs of similar nuclei can be used to test various theoretical predictions because the ratio of their respective matrix elements should be near unity. In the case of Dirac decay, the ratio  $\rho_{2\nu} = {}^{128}\lambda_{2\nu}/{}^{130}\lambda_{2\nu}$  is much smaller than the Majorana decay ratio  $\rho_{o\nu} = {}^{128}\lambda_{o\nu}/{}^{130}\lambda_{o\nu}$ , where  $\lambda$  is the probability of double beta decay. Thus even a small Majorana decay contribution would cause a drastic increase in the measured ratio  $\rho_{\Sigma} = {}^{128}\lambda_{\Sigma}/{}^{130}\lambda_{\Sigma}$ above the base level of Dirac decay  $\rho_{2\nu}$ , (where  $\Sigma$  represents the sum of the double beta decay mode components).

The measurement of the half-life of the double betadecay of <sup>128</sup>Te is far more difficult than for <sup>130</sup>Te because the probability of double beta decay is so much smaller. However, a recent measurement of the double beta decay half lives of <sup>128</sup>Te and <sup>130</sup>Te (Bernatowicz *et al.* 1993), has determined the daughter product ratio <sup>128</sup>Xe/<sup>130</sup>Xe with greater confidence than previous measurements listed in Table 3 (Manuel 1991). The mean value for the ratio of half-lives in Table 3 is 4 10<sup>-4</sup>, which compares favourably with the Bernatowicz *et al* (1993) value of 3.52 (±0.11) 10<sup>-4</sup>. The data of Bernatowicz *et al* (1993) give a limit to the effective Majorana mass of the neutrino of 1.5eV.

#### Table 3

Values reported for the ratio of the double-beta decay rate of <sup>128</sup>Te relative to that of <sup>130</sup>Te (Manuel 1991)

Reference	$^{130}\mathrm{T_{1/2}}^{/128}\mathrm{T_{1/2}}^{128}$		
Hennecke et al. (1975)			
Kirsten (1983)	1.0 10-4		
Kirsten et al. (1986)	1.0 10-4		
Manuel (1986)	5.0 10-4		
Takaoka & Sagawa (1988)	<6 10-4		
Lin et al. (1988)	3.9 10-4		
Takaoka & Sagawa (1990)	3.2 10-4		
Lee et al. (1990)	4.3 10-4		
Current best estimate	4 10 <sup>-4</sup>		

It took 20 years from the time that Pauli first suggested the existence of a neutrino to when Inghram & Reynolds (1949) undertook a determination of the halflife of double beta decay. However, a further 44 years were to elapse before the half-life of double beta decay was measured by solid source mass spectrometry. Improvements in thermal ionisation mass spectrometry have occurred to such an extent over the past decade that it is now possible to conceive of the measurement of the half-life of double beta decay schemes which have a nongaseous daughter product.

In fact, Kawashima *et al.* (1993) have recently measured the half-life of the double beta decay of <sup>96</sup>Zr from

the excess amount of <sup>96</sup>Mo found in old zircon samples. A solid source mass spectrometer was adapted to improve the sensitivity for Mo isotopic analyses by the use of ion enhancing media. The resulting half-life for the double beta decay of  ${}^{96}$ Zr of 3.9 (± 0.9) 10<sup>19</sup> y is lower than the measured half-lives of  ${}^{130}$ Te and  ${}^{82}$ Se. Zircons have a particularly tight lattice structure and this property has been successfully exploited in U/Pb geochronology because the radiogenic Pb has remained in-situ over extensive geological periods. Unfortunately the presence of U in the zircon samples generate spontaneous fission product Mo which make the measurement of double beta decay <sup>96</sup>Mo difficult to identify. Kawashima et al. (1993) were forced to assume that the spontaneous fission yields for <sup>238</sup>U (which so far have not been measured) are identical to those of the thermal neutron fission of <sup>239</sup>Pu to make the necessary corrections. The authors also report a "strange excess" in <sup>98</sup>Mo which they were unable to explain. However, despite these difficulties the work of Kawashima et al. (1993) has shown that isotopic systems, other than those with gaseous daughters, can be used to measure the half-life for double beta decay.

An examination has been made of the various double beta decay schemes listed by Doi et al. (1985) which have non-gaseous daughters. The best candidate seems to be the decay of  ${}^{122,124}$ Sn  $\rightarrow {}^{122,124}$ Te. This decay scheme has the advantage that two isotopes can experience double beta decay, old cassiterite samples of known age are available, and the mass spectrometry of tellurium is well established (Loss et al. 1990). Furthermore, this system has the great advantage with respect to the Zr/Mo double beta decay system in that cassiterite does not contain a significant amount of uranium, and in any event tellurium is close to the mass valley of nuclear fission where the cumulative fission yields are very low. In addition, cassiterite samples can be converted to the metallic form (McNaughton & Rosman 1991), and ion exchange chemical techniques have been established to separate tellurium from other elements (Loss et al. 1990). Doi et al. (1985) predict a half-life for <sup>124</sup>Sn which is approximately a factor of ten longer than that predicted for <sup>96</sup>Zr double beta decay.

It is possible that accelerator mass spectrometry could be applied to the problem of measuring the half-life of double beta decay systems because of this technique's ability to remove molecular interferences and the separation of isobars by rate of energy loss or range measurements. This enables high sensitivity mass spectrometric measurements to be made as an alternative to conventional solid source mass spectrometry, although the necessity for precise determinations of the isotopic composition of the element concerned may present a problem.

## Conclusions

It seems surprising that 65 years after Pauli's postulate of the existence of the neutrino, we still do not know some of its basic properties. Neutrinos were produced in huge numbers during the very early stages of the Big Bang, but because of their extremely low interaction with other particles they seem destined to inhabit the Universe as lonely "ghosts of creation". They are also produced copiously from every Main Sequence star, and play an essential role in supernovae explosions (Clayton 1989). Some mass spectrometric evidence to support this latter phenomenon has recently been published (Maas *et al.* 1996). It is a strange quirk of Nature that double beta decay, which is an extremely rare nuclear phenomenon, may be the mechanism which will enable the properties of the neutrino to be better understood. The basic instability of certain r-process nuclides contrasts with their present abundance distribution, which can only be explained if double beta decay rates have an extremely small probability.

The neutrino is the only fermion for which we do not know if it is different from its antiparticle and is therefore a Dirac particle, or if it is identical with its antiparticle and therefore a Majorana particle. Double beta decay offers a novel means of testing Grand Unified Theories of particle physics. In double beta decay two modes are possible - one in which two antineutrinos accompany the emission of two beta particles (the Dirac neutrino) and the second in which no neutrinos are emitted. The clear signature of neutrinoless double beta decay is the emission of two electrons whose combined kinetic energy equals the mass difference between the parent and daughter nuclides. If the latter mode can be experimentally established, it may yield limits on the electron neutrino mass, indicate the degree of lepton conservation in elementary particle interactions and provide fresh evidence for neutrino oscillations. The large difference in half-life between the two modes of double beta decay offers a sensitive indicator of estimating the contribution of neutrinoless decay to the well established two neutrino decay mode.

A considerable body of experimental evidence for the half-life of double beta decay has now been established by both direct counting techniques and geochemical measurements, although the latter technique probably provides an upper limit because of the high probability of gas loss from old tellurium and selenium-rich minerals. However, new geochemical methods based on the detection of solid daughter products by sensitive solid source mass spectrometric techniques should resolve the gas loss problem. Existing evidence suggests that neutrinoless double beta decay does not occur to any significant extent, that the electron neutrino mass may be as small as 1.5eV and that lepton conservation is maintained. However the evidence is fragmentary and plagued by experimental difficulties so that further work, especially on double beta decay, is required.

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