CAN ENVIRONMENTAL FACTORS AFFECT HALF-LIFE IN BETA-DECAY? AN ANALYSIS

A Dissertation

by

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ABSTRACT

Early in the history of the field of nuclear science, experiments were performed to ascertain whether the half-lives of the radioactive substances being studied - isotopes then called the “radium emanation” [^{222}Rn], “radium A” [^{218}Po], “radium B” [^{214}Pb], and “radium C” [^{214}Bi] - were dependent upon any external factors. At that time, the external factors deemed most likely to affect half-life were temperature and pressure. After several experiments, designed to pick up any change in half-life in the course of changing temperature or pressure, had failed to find any significant changes, it was concluded that half-life does not depend on the physical properties of external environment. And that was the state of the field for a long time - for almost 100 years, in fact.

Fairly recently, however, half-life measurements were recorded, and published, that seemed to show a change in half-life at the few percent level for certain radioactive nuclides which were exposed to extremes of temperature - thus challenging the long-held belief in the unchangeability of half-lives. In addition to half-life changes caused by temperature change, other experiments seemed to find half-life changes caused by other external influences, including the chemical environment of the decaying radioactive nuclide, and even the distance between Earth and the Sun at the time of the half-life measurement.

In this study we present evidence that the initial beliefs in the immutability of radioactive half-life (with the exception of a few nuclides decaying by electron capture whose orbital electrons are involved in both the decay and also in the chemical bonding of those nuclides) is indeed correct; we have done this by performing precise half-life measurements on the β^- emitter ^{198}Au, the EC emitter ^{97}Ru, and on the

β^- emitter ^{198}Au when sited in gold(III) oxide, Au_2O_3, (an insulator for practical purposes). We have performed various experiments designed to detect any half-life change at the level of a few parts in 10^4 due to change in temperature, physical environment, or the Earth-Sun distance. In these experiments, we have found no significant half-life change due to any of these external factors. These results represent the most accurate demonstrations of the immutability of radioactive half-life change ever made.

To my parents ... to my wife

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1. INTRODUCTION

During its infancy, the new discipline of nuclear science faced major issues as it endeavored to establish its working principles. Early on, one of the issues being dealt with was the question of whether the radioactive half-lives of the newly-discovered substances, such as the so-called "radium emanation" (now known as ^{222}Rn) were changeable, or whether they were constant. It was also necessary for the nuclear pioneers to determine how much of an effect, if any, changes in external environment might have on the half-life for each decaying nuclide.

For example, it was not certain whether changing such basic parameters as the temperature, or the pressure of the isotope's surroundings, might alter its half-life. And there were other possibilities, such as the possibility of changing the chemical composition of the nuclide.

After a number of years of imaginative and remarkably thorough research, performed by a number of scientists including but not limited to Earnest Rutherford and Pierre and Marie Curie, a point was reached in which it was accepted by the scientific community, as a result of these experiments, that with the exception of certain radioactive nuclides, whose radioactive decay involved the interaction of the decaying nucleus with its own electron cloud [1], radioactive half-lives were (1) constant and (2) independent of changes in external environment.

This view held sway until after the advent of the 21st century, at which time certain results began to issue forth from certain research groups, especially the Rolfs' group in Bochum, Germany [2, 3, 4, 5], that observations were being made of half-life changes which were caused by extreme changes in temperature. These changes in half-life were reported to have taken place for various modes of decay, such as β^--

decay, β^+-decay, and electron capture (EC). Half-life changes were claimed that had purportedly been caused by dropping the environmental temperature of the nuclide to temperatures approaching absolute zero. Much was made of these claims initially due among other things to the possibility of this discovery being potentially useful with regard to the disposal of radioactive waste [6].

However, if these claims of changes in half-life had ultimately been substantiated, major difficulties would have arisen with regard to the recording of half-life data. For example, if half-life values were truly dependent upon the decaying isotope's surrounding temperature, pressure, or perhaps other parameters, then it would have become necessary for each research group involved in half-life study to record not only their half-life results, but also the environmental temperature, pressure, and possible other values for each half-life measurement. All this data would then have had to have been factored in to the determination of accepted values for radioactive half-lives such as those produced by the National Nuclear Data Center.

From the standpoint of our own research group, such claims of temperature-dependence for half-life value would have been disastrous. Our main concern was with the half-lives of superallowed $0^+ \rightarrow 0^+$ β^+ emitters, which are essential to fundamental tests of the standard model [7]. Their precision has typically been quoted to less than 0.05%, well below the temperature and host-material dependence claimed by the measurements in Refs. [3, 4, 5]. So much of what has already been accomplished with regard to work on *ft* values and on the Standard Model would have been thrown into doubt.

It was for this reason that we decided to go ahead with a study of the vaidity of these claims of change in half-life. We determined to put these claims to the test by using highly precise methods of measuring half-lives under the conditions of changing environmental parameters. As shall be seen in the pages that follow, we believe that we have re-affirmed the long-held belief that, with the exception of certain special cases, such as the EC-decay of ^{7}Be, as mentioned above, radioactive half-lives are indeed constant and independent of changes in external environment.

We have touched briefly on the historical aspect of this interesting and thought-provoking issue. Let us now examine in more detail what, exactly, the historical basis for these issues is, and, at the same time, let us gain an understanding of the theoretical ideas that underly them.

2. HISTORICAL OVERVIEW[1]

2.1 Environmental effects on radioactive decay rates

One of the fundamental assumptions of nuclear physics, from very early in its history, has been that nuclear half-lives do not depend on external environment. In the recent past, however, various articles have been published that have claimed to find half-life changes, due to change in temperature, change in chemical environment, and due to annual variations in the Earth-Sun distance.

In the early days of nuclear science, much work was expended in an effort to find out whether radioactive half-lives might be affected by changes in external environment. As Emery [8] states, "Workers tried to change the decay constants of various members of the natural radioactive series by varying the temperature between 24 K and 1280 K, by applying pressure of up to 2000 atm., by taking sources down into mines and up to the Jungfraujoch, by applying magnetic fields of up to 83,000 Gauss, by whirling sources in centrifuges, and by many other ingenious techniques. ... Especially interesting for its precision is the experiment of Curie and Kammerlingh Onnes [9], who reported that lowering the temperature of a radium preparation to the boiling point of liquid hydrogen changed its activity, and thus its decay constant, by less than about 0.05%. Especially dramatic was an experiment of Rutherford and

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[1] Reprinted with permissions from "The half-life of ^{198}Au: High-precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob and J. C. Hardy, 2007. The European Physical Journal A, **34**, 271-274, Copyright 2007 by European Physical Journal; "Half-life of the electron-capture decay of ^{97}Ru : Precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob, and J. C. Hardy, 2009. Physical Review C, **80**, 044501, Copyright 2009 by Physical Review C; "Measurement of the half-life of ^{198}Au in a non-metal: High-precision measurement shows no host-material dependence" by J. R. Goodwin, N. Nica, V. E. Iacob, A. Dibidad and J. C. Hardy, 2010. Physical Review C, **82**, 044320, Copyright 2010 by Physical Review C; and "Do radioactive half-lives vary with the Earth-to-Sun distance?" by J. C. Hardy, J. R. Goodwin and V. E. Iacob, 2012. Applied Radiation and Isotopes, **70**, 1931-1933, Copyright 2012 by Applied Radiation and Isotopes.

Petavel [10], who put a sample of radium emanation inside a steel-encased cordite bomb. Even though temperatures of 2500° C and pressures of 1000 atm. were estimated to have occurred during the explosion, no discontinuity in the activity of the sample was observed."

An important statement made early on, as regards the independence of radioactive decay rate with respect to temperature, was given in an important paper by M. Curie and H. Kammerlingh Onnes [9, 11] mentioned by Emery above, and published in 1913. In this work, the authors trace the brief history of the attempts, to that time, to find the proper relationship between temperature (and to some extent pressure) and radioactive half-life. They state in their article that "P. Curie has shown that the law of transformation for the emanation does not change at a temperature of 450° C nor at the temperature of liquid air" [12] [The "radium emanation" mentioned here is ^{222}Rn. Liquid nitrogen boils at 77 K and liquid oxygen at 90 K. We can assume that the referred-to temperature here was in that range]. They further state that "Various observers have proved that the penetrating radiation of radium and uranium have the same value at ordinary temperature and at the temperature of liquid air" (here the authors cite Becquerel, Curie, Dewar, and Rutherford without giving a specific reference). And "The influence of high temperatures on the radium emanation and its transformation products, particularly radium C, has also been the subject of various investigations. The results have given rise to differences of opinion. Nevertheless it would seem to be justifiable to conclude that the dependence upon temperature which was observed in some cases must be attributed to secondary phenomena of less importance, and that the radioactive constants of the above substances are not appreciably altered when the temperature is raised to 1500°" [9, 11] (here the authors cite [13]).

Curie and Kammerlingh Onnes conclude "It would have been desirable to have made a greater number of experiments and to continue these during a greater length of time; nevertheless it would appear to be justifiable even now to state that cooling of radium down to the temperature of liquid hydrogen (about 20.3° C absolute) during a period of not more than $1\frac{1}{2}$ hours does not cause a change in the gamma radiation of 1 in 1000 and probably not even of 1 in 5000. It is thus probable paying due regard to the degree of accuracy attained, that this decrease of temperature has no immediate or quickly discernable influence upon the emanation or the active deposits of short period (radium A, B and C) [note: radium A is now known to be ^{218}Po ($t_{1/2} = 3.1$ m), radium B is ^{214}Pb ($t_{1/2} = 27$ m), and radium C is ^{214}Bi ($t_{1/2} = 19.9$ m)]. But in the experiments there was no opportunity for detecting an eventual effect upon the radium itself, or a slowly developing effect upon its evolution products."

After a brief discussion of some additional experiments regarding polonium, the authors further state that "All these experiments which unfortunately are not so complete as we could have wished, confirm the independence of the radiation from the temperature, over a larger range of temperatures than had heretofore been done. Moreover these experiments have brought to light sources of error which must be taken into account, if one wants to make very accurate measurements at low temperatures."

Whether the stated accuracy of the experiment by M. Curie and W. Kammerling-Onnes [9] is justified is a matter of debate. The experiment itself was in principle very precise, having been ingeniously designed and very meticulously carried out; but the stability of the electronics is uncertain by today's standards, and it was measured with an imprecise source, in the sense that there was at that time no way of knowing what the purity of the sample might have been; precise nomenclature specifying the exact nuclide used, such as is standard practice nowadays, listing both the nuclide's

Figure 2.1: The Jungfrau, at 13,642 ft. third-highest mountain in the Bernese Alps of Switzerland; and the site (at the Jungfraujoch, a research station situated below the Jungfrau summit at an elevation of 11,388 ft.) of early experimental work done to validate the immutability of nuclear half-lives. Radioactive samples were carried here in order to ascertain whether the high elevation would change the decay constant. Research work performed here was facilitated by the installation of a train, during the time period 1896-1912, that could carry men and equipment to this lofty location.

atomic mass as well as its elemental identification, was yet in the future. This is evidenced by the use in this article by Curie and Kammerling-Onnes of the term “Experiments with polonium”, which phrase specifies neither the precise isotope of polonium used, nor its purity. Nevertheless their work, along with the work of Pierre Curie and others, was taken by the scientific community as rather strong evidence that there is no radioactive half-life dependence upon the temperature of the decaying nuclide. [Figure 2.1 shows an interesting experimental location in which work was carried out during these early days in nuclear science.]

Ernest Rutherford would eventually conclude (in 1930) that “The value of λ (the decay constant) for any substance is a characteristic constantly independent of all physical and chemical conditions” [14]. It was at about this time that nuclear scientists began to accept the immutability of radioactive half-lives, with the later accepted exception of several isotopes such as ^{7}Be which decay by electron capture and are possibly susceptible to half-life changes due to their environment: their bonding electrons may influence their decay.

Texts of nuclear science written from this date have accepted that independence; an example of this is found in the classic Nuclear Chemistry textbook written by Bernard Harvey: “We have already mentioned that the radioactive decay process is a property of the individual isolated nucleus. The probability that a nucleus will decay in a certain time interval does not depend on the state of chemical combinations, the temperature, pressure, or the presence of other atoms or nuclei. (There are certain very minor exceptions to this statement when radioactive decay involves the interaction of the nucleus with its own electron cloud.) ... Pierre Curie showed that there was no detectable change in the rate when the temperature was changed from 450 to $-186°$ C [see [13]] ... A radioactive nucleus remains unchanged, perhaps for billions of years, until it decays in one sudden act.” [1].

Work on the possible correlation between temperature and radioactive half-life, seems to have been given a much lower priority after the 1920's. Correlating our work and the work done by Curie, Rutherford, Becquerel etc. - roughly one century prior - then allows us to update their results in some sense. That this update is relevant can be easily ascertained by a quick check of the spate of recent articles regarding this or that half-life anomaly. Because our work has been highly precise [to 0.04% in some measurements] we believe our work places a final exclamation point on the conclusions of the Curie paper published 99 years ago - that there is indeed no correlation between radioactive half-life and temperature, at least between 20 K and 300 K, to a verified precision of 0.04% [15].

The Curie/Kamerlingh Onnes paper states, "It would have been desirable to have made a greater number of experiments and to continue these during a greater length of time; ...." In our work, we have performed a number of experiments, with measurement times ranging from 28 to 40 days [15] - [17]. Thus, we believe we have fulfilled the desire of the authors of the 1913 paper.

2.2 Summary of previous work

2.2.1 Survey by Emery to about 1972

G. T. Emery, in his 1972 survey [8] discusses the legitimacy of various possible perturbations of decay rates as researched up to that time. This paper is important in that it summarizes the work having been done in the field, up to about 1972. In this paper, Emery analyzes cases where half-life perturbation had been claimed. He examined the electron capture (EC) decay of ^{7}Be ($t_{1/2}$ 53.3 d) and ^{89}Zr ($t_{1/2}$ 78.4 h); and the internal conversion (IC) decays of ^{90}Nb (populated from the decay of ^{90}Mo, ($t_{1/2}$ 5.7 h)), ^{99m}Tc ($t_{1/2}$ 6.0 h), ^{125m}Te ($t_{1/2}$ 58 d), ^{169}Tm ($t_{1/2}$ 4 nsec), ^{193m}Pt

($t_{1/2}$ 4.33 d), and ^{235}U ($t_{1/2}$ 26 m). He summarizes ways that these decay rates could be affected by changes in the chemical state of the sample, changes in pressure, superconductivity, internal electric and magnetic fields, temperature changes, and plasmas [8].

Changes to the decay rate of ^{7}Be had already been studied a great deal by the year 1972, and have continued to the present day. As Lieser stated in 2001 [18], "As already mentioned, changes of transmutation properties are observed if electrons of the electron shell are involved in the transmutations, as in the case of electron capture or of emission of conversion electrons. The rate of both processes depends on the electron density at the nucleus. Consequently the half-life of electron capture, and the probability of emission of conversion electrons vary to a small degree with the number and the distribution of the electrons, in particular K electrons, in the electron shell. An influence of chemical bonding on the half-life of electron capture has been measured for light nuclides such as ^{7}Be." We shall have more to say about this interesting nuclide later.

Regarding studies of the other seven nuclides treated by Emery, Tables 2.1 and 2.2 show a summary of Emery's findings. In Table 2.1 we give the studied nuclide, its decay type, any half-life changes noted, and the relevant reference(s). In general, the shifts are small, on the order of a few parts in 1,000 or less. The two exceptions are 4% shifts for ^{90}Nb (although these results did not show a consistent pattern) and a shift of the same magnitude noted for the 1.64 keV state in ^{197}Pt when measured in chloride form and compared to the metal.

We now turn to Emery's discussion of "macroscopic ways of changing the rates of electron capture and internal conversion". Issues discussed include chemical state, pressure, superconductivity, internal electric/magnetic fields, temperature and plasmas, as discussed by Emery.

Table 2.1: Summary of Emery's survey of measured half-life changes

Nuclide	Decay Type	Change in $t_{1/2}$ noted	Reference(s)
^{89}Zr	EC	Decay rate change ($\Delta\ \lambda/\lambda$) of $(8.0\pm0.3) \times 10^{-3}$	Gagneux [19]
		Decay rate change ($\Delta\ \lambda/\lambda$) of $(6.2\pm0.2) \times 10^{-3}$ (total decay)	
^{90}Nb	IC	λ(Nb met)-λ(F cmpx)=(3.6 ± 0.4) $\times10^{-2}\lambda$(F cmpx)	Cooper [20]
		λ(Nb met)-λ(F cmpx)=(3.9 ± 0.8) $\times10^{-2}\lambda$(Nb met)	Olin [21]
		No consistent pattern	Weirach [22]
^{99}Tc	IC	$\lambda(KTcO_4)$-$\lambda(Tc_2S_7)$=(2.7 ± 0.10) $\times10^{-3}\lambda(Tc_2S_7)$ (IC) (+7 state)	Bainbridge [23]
		λ(Tc)-$\lambda(Tc_2S_7)$=(0.31 ± 0.12) $\times10^{-3}\lambda(Tc_2S_7)$)(electro)	
^{125m}Te	IC	λ(Te)-$\lambda(Ag_2Te)$= $(2.59\pm0.18)\times10^{-4}\lambda$(Te)	Malliaris [24]
		$\lambda(TeO_2)$-$\lambda(Ag_2Te)$= (2.23 ± 0.18)E-4 λ(Te)	
		λ(Te)-$\lambda(TeO_2)$= $(0.36\pm0.17)\times10^{-4}\lambda$(Te)	
^{169}Tm	IC	Results of measurements shown in Table 2.2	Carlson [25]
^{193}Pt	IC	$t_{1/2}$(1.64-keV st. ^{193}Pt) in $AuCl_3$ $(4\pm2)\%$ >metal	Marelius [26]
^{235m}U	IC	λ(U)-λ(UC)=(0.318 ± 0.050) $\times10^{-2}\lambda$(U)	Shimuzu
		λ(U)-λ(USi)=(0.221 ± 0.036) $\times10^{-2}\lambda$(U)	Mazaki
		λ(USi)-λ(UC)=(0.097 ± 0.043) E-2 λ(USi)	[27, 28]

Table 2.2: Relative intensities of the P and O_1 conversion lines (from [8]) of the ^{169}Tm 8.4 keV IC transition, in different chemical environments

Probable chemical environment	P/O_1
W metal	0.056±0.007
WO_3	0.030±0.006
Tm_2O_3	0.035±0.006
Fe_2O_3	0.03±0.01

Considering first the chemical state of the decaying nuclide, he considers the results of the ^{119}Sn measurements to indicate that the valence electron density near the atomic nucleus decreases when a metallic atom forms an ionic bond. This in turn may affect the processes of electron capture and internal conversion. The length of the chemical bond can exert an effect on these processes as well, as was seen in the case of ^{99}Tc. The degree of bond ionicity, the bond length, and differences in the electronegativity of bonding partner atoms may all play a role in possible changes in EC or IC decay rates. The results given as of the date of this article by Emery are only semi-quantitative, however.

With regard to the external pressure of the decaying nuclide, it is reasonable to expect that, with increasing pressure, the densities of the valence electrons around the nucleus may become greater. This could then produce an increase in the decay rates of internal conversion and electron capture. Experiments have been done with ^{99}Tc [29], [30] and with ^{90}Nb [31], and by performing a theoretical calculation using the Thomas Fermi statistical potential, Porter and McMillan [32] have estimated the magnitude of the possible effect. They found “for the experimental pressure of 0.1 megabar a fractional decrease in lifetime of (2 to 4)$\times 10^{-4}$, the quoted variation residing in the uncertainty of the structure of the 4p band. This result agrees with Bainbridge's measurement [[33], see also [23], [34]], $(2.3\pm0.5)\times 10^{-4}$, within the accuracy of our calculation." Large increases in pressure may directly "squeeze" electronic wavefunctions, and may even produce a transfer of electrons between different bands [8].

Superconductivity: Olin and Bainbridge have demonstrated effects on IC rates of ^{99}Tc and ^{90}Nb due to superconductivity [35]. The samples were cooled to approximately 4 K and measurements taken with and without superconductivity being present. The results of this study showed a change of $0.5 \times 10^{-3}\,\lambda$ and $2.0 \times 10^{-3}\,\lambda$ for the technitium and the niobium. These effects are somewhat large; no "convincing explanation of the magnitude of the effects has been presented" according to Emery [8] in 1972.

Internal electric/magnetic fields: when atoms of ^{85}Sr, ^{89}Zr and ^{99m}Tc are substituted into the lattice of a barium titanate crystal [36], [19], [37], an "influence of the ferromagnetic phase transition of barium titanate has been observed" on the IC and EC decay rates [8], with the decay being slower in the ferroelectric phase for all three isotopes. While the effect was small for ^{85}Sr, it was measured to be $0.8 \times 10^{-3}\,\lambda$ for ^{89}Zr and $2.6 \times 10^{-3}\,\lambda$ for ^{99}Tc.

When the phase changes, from cubic paraelectric to tetragonal ferroelectric, the heavy ions undergo shifts in position, of about 0.1 $\AA$, in relation to the oxygen ions [38]; and the lattice constants of $BaTiO_3$ change [39]. "These changes in bond lengths are probably sufficient to account for the observed rate changes" [8].

Additionally, Leuenberger [40] has conducted an experiment to detect any effect caused by a condenser's dielectric field on the decay rate of ^{99m}Tc. Using fields of approximately 2×10^{4} V/cm, effects were noted of the order of 10^{-4}.

Temperature: we shall have much more to say on this subject both later in this section, and also throughout the remainder of the dissertation. However we can append this note from Emery: “Except in the vicinity of phase transitions, ... the effects of temperature changes on capture and conversion rates should be of modest size, and may be characterized in much the same way as temperature-dependent Mossbauer shifts. The volume effect, due to thermal expansion, will be similar to that found in pressure studies. There may be specific effects of changing rms vibrational amplitude, from squeezing of wavefunctions and, in polar environments, from high frequency electric fields” [8].

Plasmas: “Ionization of the atom would be a direct way of altering the rates of electron capture and internal conversion decays. Matter in stellar interiors is ionized, in general, with charge compensation by an electron gas” [8]. Bahcall [41] and Iben et al. [42] have studied ”alterations of the decay rate of ^{7}Be and the resulting changes in the expected neutrino flux”.

As we have seen, Emery provides a very thorough analysis of the work done through 1972 with regard to potential half-life change in β-decay. We should note here however that his work gives no evidence of changes to β-decay half-lives.

2.2.2 Results published since 1972

This brings our study of the survey by Emery to a close. Let us now look at the work in this area that has been done from where Emery leaves off. In 1991, O. Dragoun [43] reported a claim for a tremendous change in the ^{235m}U internal conversion half-life, following experiments in which ^{235m}U was deposited on or placed within metallic silver. These experiments were conducted by V. V. Kol’tsov and A. A. Rimskii-Korsakov [44]. This work involved the 76.8 eV E3 transition in ^{235m}U; in this case, conversion can only occur from the P or Q shells. Kol’tsov and Rimskii-Korsakov claimed that when ^{235m}U was placed in metallic silver, the half-life increased from the normal 26 m to as much as 230 m when compared with ^{235m}U atoms that had been placed upon metallic backing only (the silver having been melted off by increased temperatures).

During the experiment, the decays were followed with an electron channel multiplier. The number of counts was corrected for background for each measurement during the analysis. A total of 15 experiments were performed, and in about half these cases, the effect was observed; however, importantly these results were not reproducible. So we are left with considerable doubt as to the validity of these measurements. No follow-up work on this has been reported.

In a 1998 article, A. S. Barabash [45] considers the process of two-neutrino double β -decay. He compares the probability of this process for ^{82}Se and ^{96}Zr, and also analyzes data regarding ^{130}Te. He proposes that the probability of these events in "geochemical" measurements - those measurements that would characterize 2 β 2 ν decay probability, occurring some 10^9 years ago - is systematically lower than present-day probabilities. He then goes on to correlate this decreased probability with a change in the weak-interaction constant.

Barabash compares the rate of double β decay as obtained by counter, in modern experiments, with rates of that process in geochemical data that provide information regarding 2 β decay in past times. These latter are informed by 2 β product separation from ancient minerals, followed by isotopic analysis. After examining the available data, Barabash concludes that the discrepancies can be explained by a change, over time, of the G_F constant. The discrepancies include the following: (1) a difference in half-life values for ^{82}Se between geochemical measurements and the present; (2) the present-day half-life value of ^{96}Zr is lower than geochemical values (the error bars are such however that this result is equivocal), and (3) for ^{130}Te, (geochemical) measurements of "young" samples have lower half-life values than "old" samples.

Barabash goes on to encourage further experimentation in this area, including performing work on the current values of the 2 β (2ν) decays for ^{82}Se, ^{96}Zr and ^{130}Te. He claims that further studies are necessary to reliably and definitely confirm the validity of these discrepancies.

^{7}Be is a nuclide that has been well-known for some time to show anomalies, and a large amount of work has been put forth in studying it. This nuclide decays via EC with a half-life of roughly 53 days. Here we look briefly at some of this research.

In 1969, H. W. Johlige *et al.* studied the decay rate of ^{7}Be in various chemical combinations [46]. These combinations included BeO, BeF_2, $Be_4O(CH_3COO)_6$, $BeBr_2$, $Be(C_5H_5)_2$, and $Be^{2+}(OH_2)_4$. Their method consisted of measuring the decay constant, for various pairs with different chemical composition of beryllium, using a differential-ionization-chamber technique. The results are shown in Table 2.3.

The authors claim that the decay constant for ^{7}Be is proportional to the density of the electron cloud at the ^{7}Be nucleus. They conclude that this electron density is determined by two factors, the electronegativity of the compound in question, and the compound's lattice structure. From the experimental results, the decay constants for the various ^{7}Be compounds can be listed in decreasing order, as follows: λ $(Be_4O(CH_3COO)_6) > \lambda\ (Be^{2+}(OH_2)_4) > \lambda\ (Be) > \lambda(BeO) > \lambda\ (Be(C_5H_5)_2) > \lambda\ (BeF_2) > \lambda\ (BeBr_2)$.

In 1999, A. Ray *et al.* [47] published an article in which they claimed to observe a significant change in the decay rate for ^{7}Be, depending on whether the ^{7}Be was placed in a medium of gold metal, or in Al_2O_3. Their claim was that the ^{7}Be implanted in gold metal had a lower decay rate than that placed in Al_2O_3 by 0.72 ± 0.07 %.

After measuring this change in decay rate, the authors went on to analyze their results, using Hartree calculations, as well as the muffin-tin orbital method. As a result of these analyses, they found that, by using a combination of the geometry and the electron affinity of the atomic lattice, they could find an “effective” electron affinity (E_{EA}) for the lattice, which was considered to be responsible for the decay-rate change. As the properties of electron affinity and electronegativity are closely related, it is interesting to compare the conclusion of these authors with that of the preceding authors.

Also in 1999, Chih-An Huh [48] performed an experiment on ^{7}Be in which he was able to improve the accuracy of the Johlige results by one order of magnitude. Using a 100% efficiency high-purity germanium detector γ spectroscopy system, Huh measured the ^{7}Be decay constants for Be in BeO, $Be(OH)_2$ and $Be^{2+}(OH_2)_4$. His measurements, claiming a precision of 0.01%, yielded half-lives of 54.23 d (BeO), 53.42 d ($Be(OH)_2$), and 53.69 d ($Be^{2+}(OH_2)_4$), an observed difference of 1.5% between the largest and smallest values. This work pointed up the possibility of the ^{7}Be half-life varying significantly in natural environments. Huh explains the variation in half-life as being likely due to changes in the electron density surrounding the ^{7}Be nucleus, as Johlige had claimed previously.

As we continue to briefly survey some of the work that has been done on possible half-life changes in ^{7}Be, we next come to work that was performed by Zhi-Yi Liu *et al.* [49] in 2002. Liu *et al.* measured the decay rate of ^{7}Be implanted into a gold host and into a (natural) beryllium host. Since there is a large difference in the electron affinities of gold and beryllium, a fairly large change in the ^{7}Be decay-rate was expected, on the order of 0.7%. However, the actual measurement showed that there was surprisingly no change in the decay rate, within the limits of the precision of the experiment, which was 0.12%. From this, the authors concluded that in addition to the relative electron affinities of the ^{7}Be-containing substances being measured, the lattice structure also should be considered - as had been mentioned by Johlige earlier on.

The half-life of ^{7}Be has also been studied when the ^{7}Be nuclide is placed inside encapsulated cages composed of carbon atoms. In 2004, T. Ohtsuki *et al.* [50] measured the ^{7}Be half-life with the ^{7}Be sited in beryllium metal, and also in C_{60} "cages". Their results showed a half-life difference of 0.83%, with the ^{7}Be-in-metal half-life being 53.12±0.05 days, and the ^{7}Be sited in C_{60} half-life being 52.68±0.05 days.

It is noted within their article that the chemical environment within the C_{60} cages is altered by the presence of extensive π-bonding between the various carbon atoms. This electron-rich environment should affect the electron density at the beryllium nucleus, which in turn produces the observed change in half-life.

A. Ray *et al.* [51] in 2006 performed an interesting study in which ^{7}Be nuclides were placed both inside fullerene compounds (C_{60} compounds) and also outside them. The authors found that the half-life of ^{7}Be when sited within the fullerene (endohedral) was shorter by more than 1% than for ^{7}Be sited outside the cage. They also found that "exohedral" ^{7}Be exhibited a half-life the same as for ^{7}Be when implanted in gold metal, within 0.2%.

In analyzing their results using the linear muffin-tin orbital method calculations, the authors discovered another interesting point - when remaining outside the fullerene cages, the ^{7}Be atoms tended to move to one of the faces of the C_{60} lattice and to remain approximately $5.3\mathring{A}$ from the center of the cage, while those ^{7}Be atoms that entered the C_{60} cages seemed to move to the centers of the cages.

Next, in 2007 [52] Y. Nir-El *et al.* performed a study on the ^{7}Be half-life when that nuclide was implanted in four different materials: polyvinyl chloride (PVC) $(C_2H_3Cl)_n$, copper, aluminum oxide (Al_2O_3) and aluminum. Their results are shown in Table 2.4.

As can be seen from the table, these results are fairly consistent. They are in fact “consistent with a null host dependence within two standard deviations” [52]. On closer inspection, however, use of a statistical test showed a slight trend of half-life versus electron affinity, with conductor host-materials showing longer half-lives than insulating materials. The authors conclude “the present results may exhibit a slight (0.22%) increase of the half-life at room temperature for metals compared to insulators that requires further studies” [52].

Recently the β-decay of stored, highly-charged ions has been studied at GSI in Darmstadt, Germany (see [53]). According to the authors, “Bound-state β^--decay enhances the neutral-atom Q-value roughly by the binding energy of the generated bound electron. If this “saved” energy enbles the transition to a nuclear state of the daughter ion with much more favorable selection rules for the involved angular momenta of nucleons and leptons, the decay probability can increase tremendously. This has been proved for the β^- decay of bare $^{187}\mathrm{Re}^{75+}$ nuclei, where an enhancement of the β-decay probability - with respect to neutral $^{187}\mathrm{Re}$ atoms - by more than 9 orders of magnitude has been observed” [53]. More studies are planned for the future.

2.2.3 Claims for electron-screening effects

We start this section with a brief discussion of the concept of electron screening. The concept of electron screening (also called electron shielding) is usually encountered in an introductory chemistry course, and involves the shielding of electrons in outer, more distant electron shells, from the attractive power of the positively-charged protons in the nucleus, by the more low-lying electron shells that are situated between the outer electrons, and the nucleus. The concept of electron shielding is important in the discussion of such atomic properties as electron affinity, ionization potential, and electronegativity.

The probability that a particular nuclear reaction will occur is referred to as that reaction's cross-section. In 1987, H. J. Assenbaum and K. Langanke [54] studied cross sections (referred to as σ(E)) at low energies using the Born-Oppenheimer approximation method and a simplified model. Their studies indicated that the cross sections for certain nuclear fusion reactions, of interest to astrophysicists, were enhanced by the presence of an electron cloud, and that the enhancement could potentially be seen at standard beam energies. Since these energies were readily obtainable, the authors suggested experimental follow-up work to determine whether the effect would be observed empirically.

For a nuclear fusion reaction induced by a charged particle, at energies that are well under the Coulomb barrier, Assenbaum and Langanke propose that the cross section of the reaction drops significantly. In the context of astrophysics, the cross section value can be replaced by a different function, which is referred to as an astrophysical factor (S(E)). This astrophysical factor is defined as follows:

$$\sigma(E) = S(E)E^{-1}exp(-2\pi\eta) \tag{2.1}$$

Here, with Z_1 and Z_2 being the charge numbers of the nuclei interacting,

$$2\pi\eta = 2\pi Z_1 Z_2 e^2/\hbar\nu = 31.29 Z_1 Z_2 (\mu/E)^{1/2} \tag{2.2}$$

is referred to as the Sommerfield parameter. Mu (μ) is the reduced mass, in amu, E is the center-of-mass energy, in keV.

For a positively-charged projectile, the target nucleus is effectively screened for the incoming projectile by the electron cloud which surrounds the nucleus. The positively-charged projectile must cross the atomic radius in order to begin to feel repulsion. The amount of shielding can be crudely estimated by the ratio R_n/R_a, where R_n is the nuclear radius and R_a is the atomic radius. The ratio is of the order of 10^{-5} [54]. For most reactions, this value would be negligible; however, when the reaction involves very low energies, the shielding value becomes important: "... the penetration through a shielded Coulomb barrier at projectile energy E is equivalent to that of bare nuclei at energy $E_{eff} = E + U_e$ [note: U_e signifies the electron screening energy]. The shielding effect reduces the Coulomb barrier and increases the penetration of the Coulomb barrier. Thus, it increases the cross sections of nuclear fusion reactions .... at energies $E/U_e \leq 100$ the shielding effects cannot be disregarded and become important for the understanding of the low-energy data" [54].

The screening energy (in eV) is defined as

$$U_e = (e^2/4\pi\epsilon_0)Z_1Z_2(n_{eff}\rho_a/T)^{1/2} \tag{2.3}$$

where T is the temperature of free electrons in units of K, n_{eff} is the number of these electrons per metallic atom, ρ_a is the atomic density in atoms/m^2, and Z_1, Z_2 refer to the incident, target atoms.

With this definition, we can further define a term f_{lab}, called an "enhancement factor", as

$$f_{lab} = \sigma_s(E)/\sigma_b(E) \simeq exp(\pi\eta U_e/E) \tag{2.4}$$

where $\sigma_b(E)$ represents the reaction cross-section for bare nuclei, $\sigma_s(E)$ represents the (higher) cross-section for screened nuclei, η is the Sommerfeld parameter and E is the center-of-mass energy.

In 1994, U. Greife *et al.* studied two fusion reactions, the $d(d,p)t$ and $d(d,n)^3$He reactions [55]. He demonstrated that at energies of less than 10 keV, the cross section for the *d(d,p)t* reaction was significantly enhanced (we shall discuss both the preceeding publications in more detail below). This measurement thus showed clear evidence of screening effects.

Greife *et al.* provide a survey and analysis of experimental work done at the Dynamitron Tandem Laboratorium, at the Rühr-Universität Bochum, and at LUNA (Laboratory Underground for Nuclear Astrophysics), on (d,d) reactions. They find "the electron clouds surrounding the interacting nuclides act as a screening potential: the projectile effectively sees a reduced Coulomb barrier. This in turn leads to a higher cross section than would be the case for bare nuclei. ... For $E/U_e \leq 100$, shielding effects become important for understanding and extrapolating low-energy data" [55]. The authors conclude: "The present low-energy data clearly deviate from the trend expected from the high-energy data and can be explained by the effects of electron screening ... Although the electron screening effect has been observed for the first time in $d + d$ fusion reactions, the effect is significantly larger than can be accounted for from available atomic physics models ...." [55].

In 2002 F. Raiola *et al.* [56] reported on a study that had been performed at the 100 kV accelerator, at the Dynamitron-Tandem-Laboritorium, at the Rühr-Universität Bochum. Here, beams of deuterons were produced at energies varying from 4 to 100 keV and focused on a target of deuterated tantalum. The deuterated Ta targets were prepared by bombarding a Ta foil 0.1 mm thick with 10 keV deuterons. Four Si detectors were placed around the beam's axis, being located at a distance of 5 cm from the deuterated Ta target.

After analyzing their measurements, the authors conclude that "The electron screening effect of $d(d,p)t$ in deuterated Ta is indeed about one order of magnitude larger than in a gas target and must arise, therefore, predominantly from the deuteron environment in the Ta matrix." Thus, "the recent observation [note: this refers to [57]] of a large electron screening effect in the $d(d,p)t$ reaction using a deuterated

Ta target has been confirmed using somewhat different experimental approaches: $U_e = 309 \pm 12$ eV for the electron screening potential energy. The high U_e value arises from the environment of the deuterons in the Ta matrix, but a quantitative explanation is missing" [56].

In a follow-up study in 2004, F. Raiola *et al.* [58] provide a survey of work that had been performed to further analyze the concept of an electronic screening effect. Raiola *et al.* summarized a total of 58 cases in which deuterated insulators, deuterated metals, and deuterated semiconductors had been subjected to testing to see whether the electron screening effect had been observed.

The apparatus and experimental set-up were as mentioned for the 2002 study above [56]. Raiola *et al.* found that, out of the 58 samples studied, a large screening effect was found for most of the metals in comparison with a gaseous D_2 target. A smaller effect only, comparable to that for a gaseous target, was noted for deuterated insulators and for deuterated semiconductors. The metals studied included Be, Mg, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Sr, Nb, Mo, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Ba, Ta, W, Re, Ir, Pt, Au, Tl, Pb, and Bi. The insulators studied were BeO, B, Al_2O_3, and CaO_2. The semiconductors included C, Si and Ge.

Raiola states "The metals Sn and Pb have a high U_e value, while the semiconductors C, Si and Ge have a low U_e value indicating that high U_e values are a feature of metals.... The enhanced cross-section is most likely due to electron effects of the environment of the target deuterons. ... If n_{eff} is the number of valence electrons per metallic atom which can be effectively treated as classical and quasi-free, one may apply the classical plasma theory of Debye ..." [58]. This is the first attempt by the authors, all of whom (including Raiola) were associated with the Claus Rolfs group in Bochum, Germany, to invoke the so-called Debye theory to explain their observations.

In the context of the discussion of the more recent articles that attempt to claim changes in half-life due to change in chemical environment and change in temperature, the idea of electron screening takes on a different meaning. Within the context of the series of articles first introduced by F. Raiola *et al.* [2], electron screening is considered to be also affected by conduction electrons that are present in metallic substances. These conduction electrons are loosely held by the metallic nuclei, and are to some extent free to travel around within the metal, with each metallic atom contributing on average one conduction electron (the overall charge in the metal remains neutral).

For the purposes of the Raiola article and other similar articles along the same vein, these conduction electrons, present in any normal metal, are considered to constitute a sort of plasma, referred to by these authors as a “Debye” plasma. Because of the presence of this “Debye” plasma, it is considered that the processes involved in β-decay - i.e. β^--decay, β^+-decay, and EC - as well as α-decay are somehow either slowed down, or made to speed up, because of the presence of this “plasma”. Transitions involving the movement of a negatively-charged particle, such as an electron (β^--decay or EC) are supposed to be slowed, according to this theory. That is due to the electromagnetic repulsion between the decaying electrons, and the negatively-charged “plasma”. On the contrary, decays involving positively-charged particles (β^+-decay and α-decay) would be speeded up, since the positively-charged particle should be attracted to the negatively-charged plasma.

We also see in the Raiola article [58] the mention of the concept of temperature change being involved in the process: “A critical test of the classical Debye model is the predicted temperature dependence, $U_e \propto T^{-1/2}$.” They conclude “An explanation of the large effects in metals is possibly provided by the classical plasma screening of Debye applied to the quasi-free metallic electrons” [58].

It is contended that this “Debye plasma” alters the phase space which is available for radioactive decay, and that it either increases (for β^- or electron-capture decay) or decreases (for β^+ decay) the decaying nuclide’s half-life. This change in phase space would occur only in metals - not in insulators. Also, the phase-space change would be further enhanced were the metal to be cooled to extremely low temperatures.

We now move to a series of articles that were subsequently put forth by the above-mentioned Rolfs’ group, in rapid succession. These articles all key on the invocation of the concepts put forth by Raiola, above - the concepts of a so-called “Debye plasma”, and of the electron-screening effect in radioactive decay being affected by temperature change.

The first article in this series was produced by B. Limata *et al.* [3]. Limata studied the β^+-decay of ^{22}Na, with the ^{22}Na implanted in the metal Pd. Limata *et al.* used a setup that consisted of a cryopump, capable of lowering temperatures to 12 K, with the sample being attached to the cryopump’s head. The 511 and 1274 γ-rays emitted following the β^+ (EC) decay of ^{22}Na were monitored by a germanium detector. The measurement was performed first at room temperature (measured for two days) and then at 12 K (measured for three days).

Limata *et al.* found a difference in activity that corresponded to the half-life of ^{22}Na being shortened, by 1.2 ± 0.2 %. Since the half-life of ^{22}Na is 2.6 y, they measured the source activity in each case. They found this result to be consistent with their previous proposal of a shortened half-life for decays involving positively-charged particles. However, the magnitude of their result was smaller than that predicted by the formulas they had derived. In their derivation, the Debye potential U_D is related to U_e(d+d), the screening energy determined from the (d,d) reaction

(300 eV). From equation (2.3) they obtain

$$U_D = Z_e Z_t U_e(d+d)(293/12)^{1/2} \tag{2.5}$$

and find the equivalent enhancement factor for the decay rate to be

$$f_{lab} \approx ((Q+U_D)/Q)^5 \tag{2.6}$$

which results in a value for f_{lab} of 1.14, which indicates that the half-life should be shorter by 14%. Therefore, the decrease in half-life that was actually observed was considerably smaller than expected, according to the given equations. However the group took hope from this result, since a significant change in activity was identified, and this change corresponded to a shorter half-life, as their equations had predicted.

The second article that we shall discuss from the Rolfs' group was published by Wang *et al.* [5]. Wang studied the EC-decay half-life of ^{7}Be ($t_{1/2}$ = 53.3 d), the ^{7}Be being placed in two different metallic environments, Pd and In. The experimental setup was as in the Limata measurement given above. The Ge detector monitored the 478 keV γ-rays resulting from the EC-decay of ^{7}Be. The activity of the sample was measured at room temperature (1 - 2 days) and at 12 K, for the same time period. The $t_{1/2}$ of ^{7}Be is 53.3 d.

The formulas derived by the group to provide an estimate of the half-life change predicted that the ^{7}Be $t_{1/2}$ should have been longer at the lower temperature by 1.1%. The results of the actual measurements corresponded to a half-life increase of 0.9 ± 0.2 % in one case, when measured at 12 K in palladium, and by 0.7 ± 0.2 % in the other, when measured in indium [5], as compared to its half-life at room temperature. Again, the group considered the results to be consistent with predictions, as the results were in the same direction as the predicted half-life change, though smaller in magnitude.

The third measurement conducted by the Rolfs' group was performed by T. Spillane *et al.* [4]. This measurement involved the β^--decay of ^{198}Au. The setup was much the same as for the previous two experiments. The sample was again mounted in a cryopump, with the Ge detector being used to monitor the 412-keV γ-rays resulting from the decay. Several gold foils of thickness 0.5 mm were neutron activated at the Isotopenlabor of the Ruhr Universität Bochem. The sources were monitored for a period of about 3.5 days, a little over one half-life of ^{198}Au.

The authors reported a ^{198}Au half-life of 2.706 ± 0.019 d at room temperature and of 2.802 ± 0.020 d at a temperature of 12K. Their theory predicts that, at room temperature, the half-life of ^{198}Au located in a metal should have been 7% longer than if it had been located in an insulator. And, at 12 K the difference in half-life should have increased even further, to 32%. As reported by Spillane *et al.*, the corresponding measured results were only 0.4(7)% and 4.0(7)%, respectively. Once again, the direction of the half-life change was in the direction predicted by their model, but the magnitude in this case was much smaller than predicted. Nevertheless the authors claimed a significant dependence of the ^{198}Au half-life on temperature.

Another study by the Rolfs' group involved the α-decay of ^{210}Po ($t_{1/2} = 138$ d), sited in copper metal. This experiment was performed by F. Raiola *et al.* [2]. The setup was again as for the other measurements of the Rolfs' group, consisting of a cryopump with the sample, together with a silicon diode, installed on its cold head. The minimum temperature reached was 12 K. A silicon detector was used to detect the α-particles. For the sample, ^{209}Bi nuclides were implanted into a 99.9% pure Cu sample; the ^{209}Bi nuclides were then neutron-activated to produce ^{210}Bi. Then, the ^{210}Bi nuclides ($t_{1/2} = 5.0$ d)underwent β^--decay to ^{210}Po.

The α activity (decay rate per second) was measured first at room temperature, then at 12 K. (The duration of the measurements is not specified.) After a total of six runs, the results of the activity level at the two temperaures was compared. The result was a higher activity, by 6.3±1.0%, at 12 K. According to the authors, this result is in the expected direction but is smaller than anticipated, as determined from their calculations, by approximately a factor of 10.

To summarize: the measurements reviewed above, by the Rolfs' group, all showed a change in half-life in the predicted direction, as dictated by the formulas developed by the group. However, in all cases the change was smaller than the predicted value. All three studies suffered from being conducted over a short time period, which required the authors to rely in most cases on activity rather than direct half-life measurements. It would be greatly preferable for such a study to cover a time period many times longer than the half-life of the nuclide in question.

If such a temperature dependence, as was claimed by the Rolfs' group, could have been confirmed, it might have had a substantial impact on the way we understand nuclear β decay. If this scenario were correct, it would also lead to half-lives that were dependent upon the host - and also would, according to the authors, show a smooth dependence of half-life on the metallic temperature.

Not surprisingly the claims of these authors produced substantial popular interest. This was partly because, if true, these findings could have - potentially - helped in the disposal of radioactive waste. However, what was not commented on at the time was the potentially negative impact that such a result would have had on previous half-life measurements. All the half-lives that have ever been measured, when quoted with sub-precent precision, would have been placed in jeopardy.

2.3 Earth-Sun distance effect

Recently, researchers J. Jenkins, E. Fischbach and their co-workers, in a series of papers which commenced in 2009 and which continues to the present, have claimed the finding of systematic changes of radioactive half-lives, over a ±0.1% range, which vary as a function of the distance between the Earth and the Sun at the time of measurement (as an example, see [59]). Their claims are based primarily upon their personal interpretation of two sets of data resulting from measurements taken some time ago by other groups, at the Brookhaven National Laboratory [60] and also at the Physikalisch Technische Bundesanstalt (PTB) [61]. Jenkins *et al.* have attempted to explain the periodic variations in half-life found in those studies by claiming that they were the result of "annual variation of nuclear decay rates" [59], which are related to the annual change in the distance between the Earth and the Sun. The original authors of the measurements never made such claims themselves, although one group did state that the periodic variations could "not be fully accounted for by our tests or estimates" [60]

The Brookhaven study, conducted between 1982 and 1986, involved repeatedly measuring the activities of ^{32}Si ($t_{1/2} = 172$ y), and ^{36}Cl (a long-lived isotope with $t_{1/2}$ = 1.6 x 10^3 y) directly, and then obtaining the ratios of ^{32}Si to ^{36}Cl over a period of four years. The authors used a gas-flow proportional, end-window counter in order to detect decay β particles. Their results appear to show a correlation between the decay-rate ratios and the Earth-Sun distance, albeit with a phase shift between them.

The PTB measurements were conducted over a period of 11 years; in this study, the researchers used a high-pressure $4\pi\gamma$ ionization chamber to measure the activity of ^{226}Ra ($t_{1/2}$ = 1.6 x 10^3 y). These data also show a weak, but nonetheless statistically significant, oscillatory behavior of the decay rate, with a one-year period.

The highest rate (corresponding to the shortest half-life) occured a few days after the Earth-Sun orbit had reached its perihelion, the day and time at which the Earth and Sun are nearest one another.

When the results of these two groups were published, both groups acknowledged the oscillations present in their data. The BNL researchers noted that these oscillations corresponded with seasonal variations in temperature and in humidity, which might have affected the relative absorption of the β particles from ^{32}Si and ^{36}Cl (although they didn't measure these parameters until near the end of their work). The PTB researchers attributed the oscillations to background radioactivity, such as that from radon and its daughter products; these are known to show seasonal changes in concentration.

Despite all this, Jenkins *et al.* [59] have insisted on taking both of these sets of data exactly at face value, and have proposed causes for these oscillations that are much more basic. First, they submit the possibility that the sun might either emit a flux (see [62]), or possess a field, which varies on an annual basis. In discussing this possibility, the authors cite articles by Barrow [63] and Shaw [64] which theorize that the sun might produce a scalar field, ϕ, which could possibly cause changes in the electromagnetic fine structure constant (α_{EM}); this would then lead to a change in both α- and β-decay rates. More recently, Fischbach *et al.* [65] even theorize new particles which they refer to as ”neutrellos” could be responsible. Changes in the electron-to-proton mass ratio have also been mentioned as possible causes of the variations. Or perhaps changes in the neutrino flux produced by the sun (ϕ_ν), they speculate, might in some way affect the nuclei of radioactive elements, perhaps due to the very small magnetic dipole moment of the neutrinos. In support of this, they have published an article [66] claiming that during the solar flare of 13 December 2006, a decay-rate change of ^{54}Mn was observed. This, they contend, is “consistent with a mechanism based on a change in ϕ_ν during the solar flare" [66].

As regards the solar-flare article [66], the counting rate of ^{54}Mn was monitored by the authors over a period of roughly one month, from December 2006 to January 2007. During this time frame an earth-directed solar flare was observed, on Dec. 13. The normalized count rate for ^{54}Mn during this time was then plotted against time, with a plot showing the integrated X-ray values associated with the solar flare (GOES-11 x-ray data) being superimposed. The resulting plot shows a correlation between the increases in solar x-ray flux, on 12/12, 12/14 and 12/17, and drops in the count rate for ^{54}Mn. Jenkins et al. [66] attribute this change in the ^{54}Mn half-life to the solar flare. An additional solar flare, earlier in the month, does not correlate with a change in the ^{54}Mn count rate, but this is attributed to that solar flare's having been directed away from the earth. The authors conclude from this study that some agent (possibly neutrinos) emitted by the sun caused the drop in count rate for ^{54}Mn. If the unknown agent should prove to be the neutrino, the authors reason, the change in count rate could be accounted for, simply by a transfer of a small amount of energy - on the order of 50 eV - from the neutrinos to the decaying nucleus; i.e., it would not require a full absorption of the neutrinos for the changes in count rate to occur.

Table 2.3: Comparison of measured half-life changes (from [46]) due to changes in chemical combination for ^{7}Be.
Note that these were measurements of activity, and not half-life, directly, and are thus less precise than, for example, our own current measurements, in which half-life is directly measured.

Source Pair (λ - λ)	Result ($\times 10^{-3}\lambda$(Be))
BeO - BeF_2	1.130±0.058
BeO - $BeBr_2$	1.472±0.063
BeO - $Be(C_5H_5)_2$	0.795±0.074
$Be^{2+}(OH_2)_4$ - BeO	0.374±0.077
$Be_4O(CH_3COO)_6$ - BeO	0.724±0.057
$Be^{2+}(OH_2)_4$ - $Be(C_5H_5)_2$	1.169±0.106
$Be_4O(CH_3COO)_6$ - BeF_2	1.852±0.082

Table 2.4: Nir-El measurement half-life results for ^{7}Be implanted in four different substances [52].

Host material	$t_{1/2}$
copper	53.353(50)
aluminum	53.257(44)
Al_2O_3	53.180(43)
polyvinyl chloride	53.181(45)

3. OBJECTIVES OF THE CURRENT WORK[1]

It should be kept in mind that the most important concern regarding the possibility of weak-decay half-lives being susceptible to external environmental influences is that perhaps all previous such half-life measurements would be thrown into doubt. For us, the area of greatest concern regards the half-lives of superallowed $0^+ \rightarrow 0^+$ β^+ transitions, since these half-lives are vital to fundamental tests of the standard model [7]. The precision of these half-lives has typically been quoted to less than 0.05%, which is obviously well below both the temperature and host-material dependence that were claimed by the new measurements [2] - [5].

3.1 Temperature dependence of ^{198}Au β^--decay

Our first half-life measurement comparison involved ^{198}Au. We show its decay scheme in Fig. 3.1. After we devised a way to measure the half-life of this nuclide (and subsequently other nuclides) at very low temperature, we started our research by first measuring the half-life value for ^{198}Au at the temperature of 19 K, and then measuring the half-life value for the same nuclide at room temperature. During our measurements, we monitored the β-delayed γ ray (in ^{1998}Hg) at 412 keV.

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[1] Reprinted with permissions from "The half-life of ^{198}Au: High-precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob and J. C. Hardy, 2007. The European Physical Journal A, **34**, 271-274, Copyright 2007 by European Physical Journal; "Half-life of the electron-capture decay of ^{97}Ru : Precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob, and J. C. Hardy, 2009. Physical Review C, **80**, 044501, Copyright 2009 by Physical Review C; "Measurement of the half-life of ^{198}Au in a non-metal: High-precision measurement shows no host-material dependence" by J. R. Goodwin, N. Nica, V. E. Iacob, A. Dibidad and J. C. Hardy, 2010. Physical Review C, **82**, 044320, Copyright 2010 by Physical Review C; and "Do radioactive half-lives vary with the Earth-to-Sun distance?" by J. C. Hardy, J. R. Goodwin and V. E. Iacob, 2012. Applied Radiation and Isotopes, **70**, 1931-1933, Copyright 2012 by Applied Radiation and Isotopes.

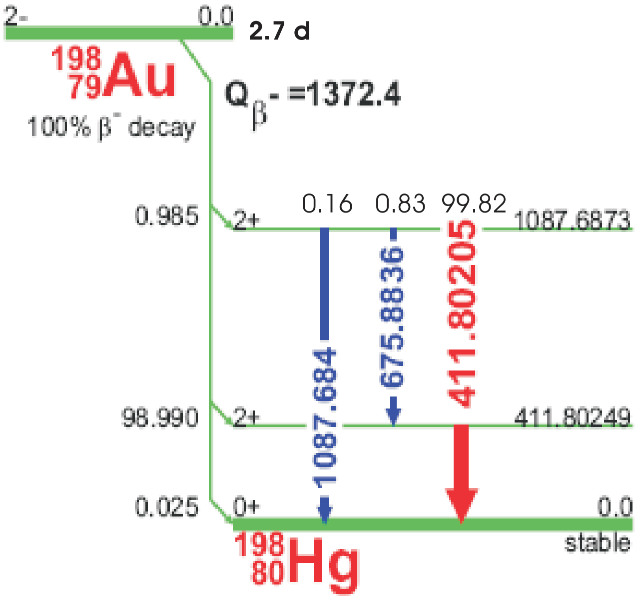

Figure 3.1: Shown here is a partial decay-scheme for the β^- decay of ^{198}Au; it shows the dominant transitions and the γ rays which follow them. This information is taken from Ref. [67]. We measured the ^{198}Au half-life by following the time decay of the β-delayed 412-keV γ ray (in ^{198}Hg).

3.2 Temperature dependence of ^{97}Ru EC-decay

We next set out to determine the temperature dependence for the EC-decay half-life of a nucleus with a Z considerably larger than that of ^{7}Be. We hoped in this to achieve a precision comparable to those that had been obtained for β^- decay, i.e. $\ll$ 0.1%. We sought a nucleus for our study that decayed entirely by electron capture, if possible, and preferably with a half-life on the order of a few days; and with a delayed γ ray that could be detected cleanly. In addition to these requirements, our nucleus also had to be capable of being produced via thermal-neutron activation, so that we could obtain statistically useful quantities without having to deal with serious contaminants. In searching for this isotope we found that, although there are not a lot of candidates from which to choose, one - ^{97}Ru - happily satisfied all our requirements. We show its decay scheme in Fig. 3.2. Since the maximum decay energy for any allowed transition from ^{90}Ru is 892 keV, the nucleus must decay purely by electron capture. We also report here measurements of the half-life of ^{97}Ru, conducted at both room temperature and at 19 K, as measured via its 216-keV β-delayed γ ray.

3.3 Temperature dependence of ^{198}Au β^--decay on host material

After our experimental work on the temperature-dependence of the β^--decay of ^{198}Au and the EC-decay of ^{97}Ru, we addressed the possibility that there might be a difference between a ^{198}Au source sited in a metal, and a ^{198}Au source sited in an insulator, as predicted by the Bochum version of “Debye” theory. We addressed this problem by conducting a measurement of the ^{198}Au half-life for which the decaying nucleii were sited in Au_2O_3.

Both measurements of the half-life of ^{198}Au in gold metal - ours [15] and that

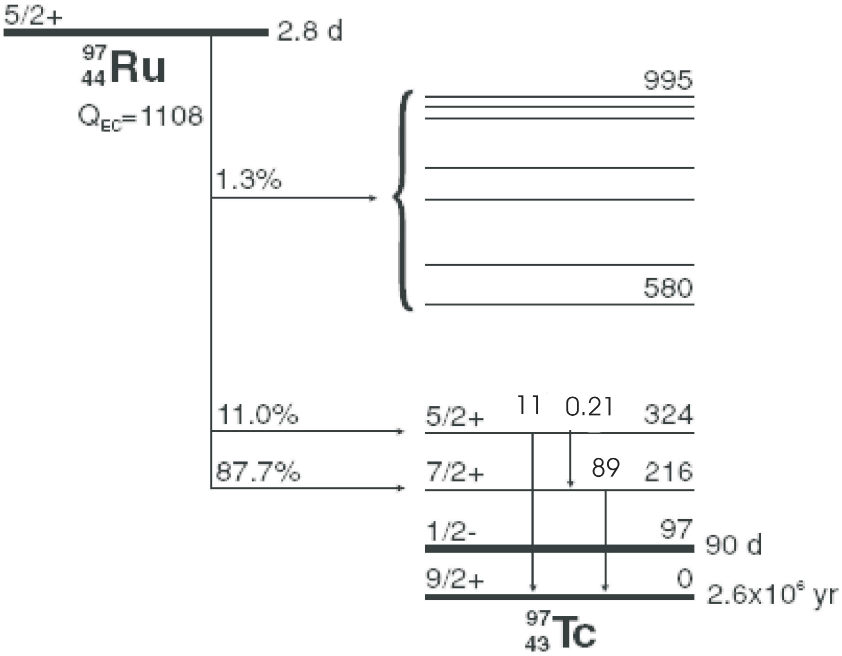

Figure 3.2: Shown here is a partial decay-scheme for the electron-capture decay of ^{97}Ru; it shows the dominant two transitions and the γ rays which follow them. This information is taken from Ref. [68]. We measured the ^{97}Ru half-life by following the time decay of the 216-keV γ ray.

of Spillane *et al.* [4] - were conducted using sources that were prepared by neutron activation of natural gold, ^{197}Au. In order to obtain conditions that were comparable, and to obtain statistics for our measurement in a nonmetal, we desired to again use neutron activation. We therefore sought a suitable gold compound that also has the property of being an insulator. Although not strictly speaking an insulator, we identified Au_2O_3 - gold(III) oxide - as a suitable candidate. It is actually considered to be a semiconductor [69] but, with a calculated band gap of greater than 0.85 eV, it ought to behave as an insulator at room temperature. And in fact, it does so, as shown by the fact that its room temperature resistivity has been measured to be five orders of magnitude, at minimum, higher than the room temperature resistivity of gold in its pure state [70]. This resistivity difference is undoubtedly large enough to ensure that a putative conduction-electron plasma will be absent.

3.4 Temperature dependence of ^{198}Au β^--decay on Earth-Sun distance

So far, all the experimental data utilized with regard to this interpretation - the results from BNL and from PTB which have been so significantly interpreted by Jenkins *et al.* [59] - have been obtained from successive measurements of the activities of radionuclides which are long-lived. However, this sort of measurement, extending for months or even years, finds itself susceptible to systematic effects and to instabilities which arise due to changes in temperature, instrumental drifts, background radiation and humidity. In order to avoid these types of difficulties, we have undertaken a very different approach by making seven individual half-life measurements, and by making these measurements of a shorter-lived radionuclide, ^{198}Au ($t_{1/2}$ = 2.7 d). The measurements, the decay of which we followed for 10 half-lives, were repeated seven times, spread out over time such that the seven measurements extend over the full range of Earth-Sun distances.

When the first of the claims by Jenkins *et al.* [59] was published, we had already made three sequential measurements of the half-life of the β decay of ^{198}Au (for the purpose described in sections III A and III C). So we decided to enlarge our study by making four more half-life measurements of ^{198}Au, to ensure that an approximately evenly-spaced set of Earth-Sun distances was being sampled. We have scattered these measurements as evenly as practicable over the perihelion-aphelion interval. If the β^{-}-decay half-life indeed depends upon the Earth-Sun distance, some change in the values of our half-life measurements should be obtained, with those changes corresponding to Earth-Sun distance changes that would have occurred during the course of our study.

Because we have depended upon direct half-life measurements of a nuclide of relatively short half-life, rather than upon separate activity measurements which were spaced over a long period of time, we have been able to substantially lower the potential effects of instrumental drifts.

Now that we have overviewed the objectives for our measurements, let us take a look at how we set about to execute the measurements. In doing this, we utilized high-precision half-life measurements to determine if the claims that had been made for half-life changes were valid. In the next section, we take a look at the experimental apparatus we used in our research.

4. EXPERIMENTAL APPARATUS[1]

In this section, we discuss the experimental set-up utilized by us as we conducted our various measurements. Our experimental set-up was designed to be as simple as possible, so as to minimize unnecessary systematic effects. A picture of the apparatus is shown in Figure 4.1.

For all our measurements, temperature-dependence, host material and Earth-Sun distance-dependence, we used substantially the same set-up. Our apparatus consisted of primarily a cryopump, in which we placed the radioactive sample, and a germanium detector which was placed such that the face of the detector was as near as possible to the sample. The basic layout of our electronic set-up is shown in Figure 4.2. A compressor/roughing pump combination was also required for the cold-temperature measurements.

We now look at the set-up for the experiments in more detail. For the temperature-dependence measurements, i.e. the ^{198}Au and the ^{97}Ru measurements, we used the same set-up -exactly- for both the room temperature and cold measurements. We utilized a CryoTorr 7 model cryopump as a cooling source. For the ^{198}Au temperature-dependence and the Earth-Sun measurements, we placed the irradiated gold samples directly on the cold head of the cryopump. For the ^{97}Ru measurements, we attached

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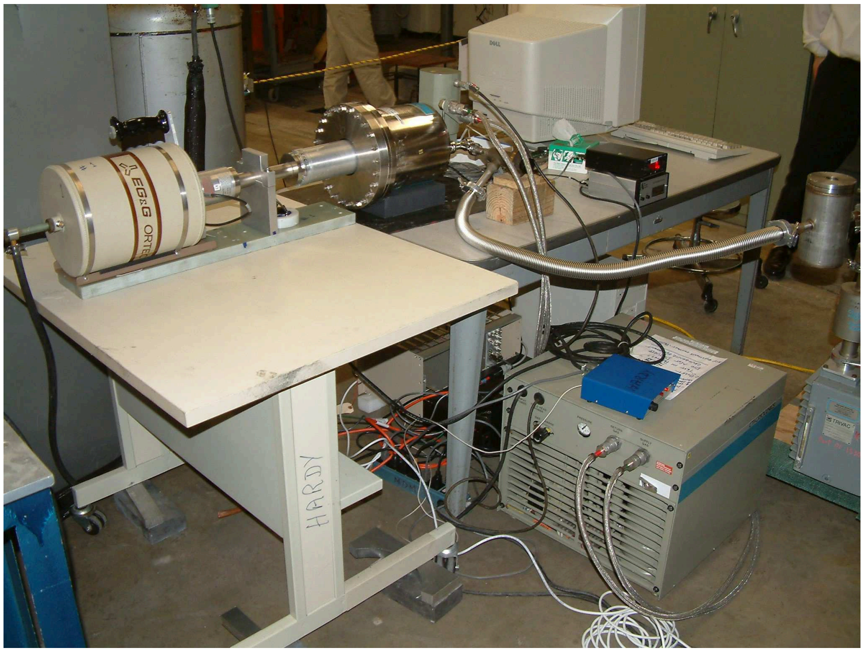

Figure 4.1: Our experimental setup for the ^{198}Au cold measurement. We used a 70% HPGe detector (shown at left, on the table), with the face of the detector positioned inside a cavity in the cryopump (center, on table), a CryoTorr 7 model. Substantially the same set-up was used throughout the series of measurements.

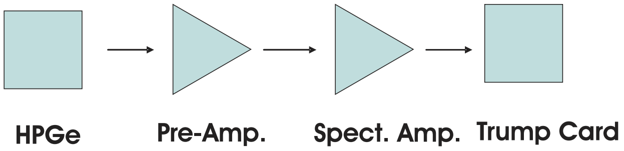

Figure 4.2: Schematic diagram for our experimental set-up for the various measurements. In the diagram, “HPGe” stands for our high-purity germanium detector, “Pre-Amp” represents our pre-amplifier, “Spect. Amp.” stands for spectroscopy amplifier, which is in turn followed by the Ortec Trump card. This same basic set-up was employed for all our experiments.

the irradiated ruthenium samples between two copper washers and then fastened the assembly directly onto the cold head of the cryopump with four symmetrically placed screws.

A 70% HPGe detector was placed so that it faced the sample on the axis of the cryopump, being located just outside the cryopump's cover plate. A cavity had been bored in but not completely through the cover plate, such that only 3.5 mm of stainless steel remained between the face of the detector and the irradiated sample. The total distance between the face of the detector and the sample was only 45 mm. In each series of measurements following a decay, this arrangement remained unchanged throughout.

For the cold-temperaure measurements, we monitored the cold head temperature with a temperature-calibrated silicon diode (Lakeshore Cryogenics DT-670) [71] which was fastened to a point alongside the gold sample, on the cold head itself. The diode was then connected to a Lakeshore Model 211 temperature monitor.

When conducting a low temperature measurement, we used a roughing pump first, to decrease the pressure to roughly 9 mtorr. We then engaged the cryopump. The cold head, to which the sample was fastened, according to the literature was expected to reach a temperature as low as 12 K; however, with our independently-installed silicon diode, we measured a temperature of between 17 K and 19 K, slightly higher than the manufacturer's published specifications had indicated.

Two condensing arrays are located on the CryoTorr cryopump. An outer 80 K array condenses water as well as hydrocarbon vapors. This helps to assure that water or oil from the pump do not accumulate on the radioactive sample during the measurement; this would cause an increasing amount of absorption over time. The outer array fastened onto the cryopump assembly outside the cold head (where the sample was located). It was thus located between the sample and the cover plate of the cryopump assembly. The inner 15 K array condenses nitrogen, oxygen and argon. This array also contains a special type of charcoal that absorbs helium, hydrogen and neon [72].

The arrangement for the corresponding room temperature measurement was identical except that the pumping and cooling steps were omitted. We utilized the cryopump in this measurement also, as a sample-holder, for consistency's sake. It may be noted that we did not alternate temperatures for a single source, but rather made a complete decay measurement at one temperature with one source at a fixed geometry. Then, starting with a fresh source, we made a similar dedicated measurement at the other temperature. Our half-life results are therefore entirely independenct of any geometrical or source differences that might have occurred between the two measurements.

The samples we used for the ^{198}Au temperature-dependence and Earth-Sun measurements were circular gold foils, having a 10 mm diameter, and a thickness of 0.1 mm. These foils were obtained from Goodfellow Corporation. According to the supplied literature, the material was 99.99+% pure, having only 5 ppm As and 5 ppm Si as identified impurities. For each measurement, a foil was activated for 10 seconds in a flux of 10^{10} neutrons/(cm^2 $\cdot$ s), in the Texas A&M Triga reactor. This activated foil was then mounted on the cold head of the cryopump.

When performing half-life measurements, we can note that gold has two important advantages as a choice in sample. First, it is monoisotopic (^{197}Au), so neutron activation produces only ^{198}Au, and secondly its decay spectrum is dominated by a single strong β-delayed γ ray at 412 keV (see Fig. 3.1). No corrections are required therefore for contaminant activities, and the peak-to-background ratio is very high.

For the host-material dependence measurement (Au_2O_3), we used a gold(III) oxide sample which was obtained from the Alfa Aesar Corporation. This was in a powder form with a purity of 99.99%. A 170-mg sample composed of this powder was attached onto an aluminum disk by adhesive Mylar tape, 56 μm thick, and the assembly was activated in a flux of $\sim 10^{10}$ neutrons/cm^2, for 10 s at the Triga reactor at Texas A&M University. The irradiated Au_2O_3 sample was then placed between two thin copper discs, and these discs, with the sample between, were then firmly attached to the cold head in such a manner that the sample could not move. Although we did not cool the Au_2O_3 sample to a low temperature in this measurement, for consistency's sake we nevertheless followed the same procedure as in the previous measurement, including the use of this cryopump as the location for our sample. Although we used Au_2O_3 as the material to be activated in this experiment, all the other aspects of the measurement were identical to those of our previous experiment [15], in which we activated pure gold. We thus were able to directly compare the ^{198}Au half-lives measured at room-temperature with two different host materials, one an insulator and the other a conductor.

For the ^{97}Ru measurement, our sample was a single crystal in the form of a circular disk, 8 mm in diameter and 1 mm thick, which was obtained from Goodfellow Corp. According to the supplier, the chemical purity of the material was 99.999%, with no identifiable impurities. For each measurement, the metal crystal was initially activated for 10 s in a flux of 10^{13} neutrons/cm s, at the Texas A&M Triga reactor. This activated crystal was then fastened directly onto the cryopump's cold head, thus ensuring good thermal contact over the entire crystal area.

For the measurements themselves, sequential γ-ray spectra were recorded from the HPGe detector. The detector signals were amplified and then sent to an analog-to-digital converter, which was an Ortec TRUMP^{TM}-8k card [73] which was in turn controlled by Maestro software that had been installed on one PC, operating under Windows-XP.

Throughout the entire duration of the measurements, our computer clock was synchronized daily against the signal broadcast by WWVB, the radio station operated by the U.S. National Institute of Standards and Technology. For both the room and low-temperature measurements on ^{198}Au, six-hour spectra were acquired sequentially for more than one month - 32 days for the room temperature measurement, and 31 days for the cold measurement. In each case, more than 120 γ-ray spectra - 251 in total - were recorded. For the ^{97}Ru temperaure-dependence measurements, at both room temperature and low temperature, six-hour spectra were acquired sequentially for approximately one month. In each case, more than 110 γ-ray specra were recorded. For the host-material dependence (Au_2O_3) measurement, 107 sequential 6-h γ-ray spectra were acquired, as for the measurements above, and recorded for a total period of 27 days - 10 half-lives of ^{198}Au. And for the dedicated Earth-Sun series of measurements, six-hour γ-ray spectra from the HPGe detector were again acquired sequentially, for more than a month from each source. At least 100 γ-ray spectra were recorded for each measurement, with a total of 786 spectra being obtained for all seven measurements on ^{198}Au.

The TRUMP^{TM} card which we employed utilizes the Gedke-Hale method [74] to make dead-time loss corrections. We kept our system dead-time below about 4% and recorded all our spectra for an identical pre-set live time. We ensured therefore that our results were essentially independent of dead-time losses.

However, at a precision level of 0.1% or better, pile-up can also become an issue, as it is unaccounted for by the Gedke-Hale method; so we carefully tested our system for residual rate-dependent effects. We measured the 662-keV γ-ray peak from a ^{137}Cs source alone, first, and then remeasured that source a number of times in the presence of a ^{133}Ba source, which was moved closer and closer to the detector so that both the dead time as well as the number of chance coincidences increased. Each measurement was also made for the same preset live time.

We then obtained from each measurement the number of counts in the 662-keV peak and, from the decrease in that number as a function of increasing dead time, we found that the fractional residual loss amounted to $5.5(2.5) \times 10^{-4}$ per 1% increase in dead time. At the count rates experienced during our ^{97}Ru measurements, the required correction was never greater than 0.2% but it was nevertheless applied to all spectra. This correction was also applied to all spectra when we performed the subsequent measurements on ^{97}Ru, Au_2O_3, and ^{198}Au in conjunction with the Earth-Sun measurements. The basic layout of the set-up we used to determine the fractional residual loss due to pile-up is shown in Figure 4.3.

We have examined our experimental set-up; let us now look at how the data we gathered was analyzed. We do this in the next section.

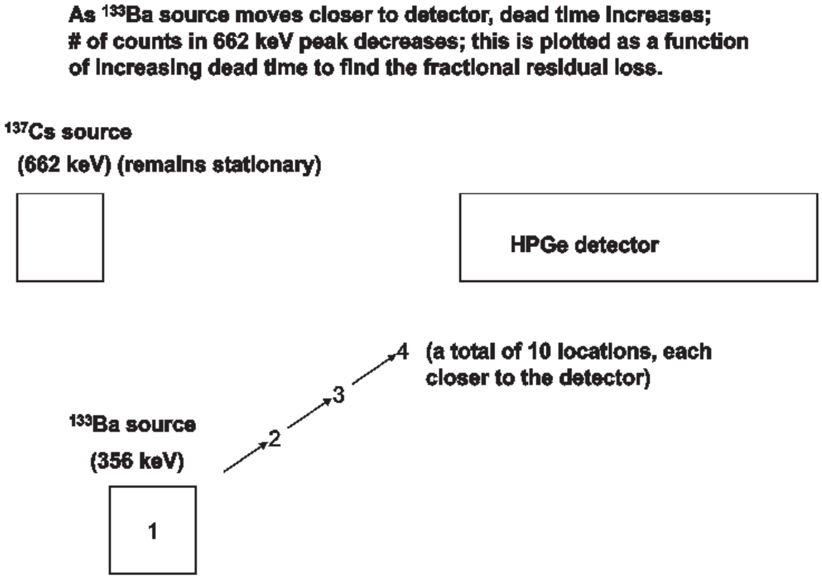

Figure 4.3: Diagram showing the experimental set-up to test for residual rate-dependent effects; see text for a discussion of our experimental procedure.

5. ANALYSIS[1]

We now turn to our experimental analysis for the measurements we have performed. For all the ^{198}Au measurements, including the two temperature-dependence measurements, the gold(III) oxide (Au_2O_3) measurement, and the dedicated Earth-Sun measurements, the prominent 412 keV γ-ray peak in each recorded spectrum was analysed with GF3, a least-square peak-fitting program in the RADware series [75]. This peak-fitting program allowed us to be very specific in determining the correct background for a peak, and the 412 keV peak in each spectrum was visually inspected to accomplish this. We applied the same criteria to each spectrum so far as possible. Fig. 5.1 shows a sample peak with its fitted background, from which its area was determined.

With Radware, there are six basic parameters that can be fixed or adjusted by the program to give the best fit to a peak. Parameters A, B, and C are used to set the background, where the background = A + B*X + C*X^2. The peak shapes are determined by the parameters STEP, R and BETA, and by P, W and H, the position, width and height of the peak. In our analysis we used primarily the parameters A, B, and STEP. The STEP parameter determines the height of a step function to account for higher background below a peak. The higher background on the low-energy side

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of a peak is due either to Compton scattering of photons outside the detector that are directed into the detector, or to electrons having escaped from the semiconductor crystal [76]. The peak itself is fitted by summing a Gaussian with a height = H*(1-R/100) and a skew gaussian with a height = H*R/100. BETA sets, in channels, the decay constant of the skew gaussian.

When a gamma photon enters a detector (such as the type used by us, a high-purity germanium detector), electrons are generated by three processes: the photo-electric effect, Compton scattering, and pair production; the electrons thus produced are then detected [77]. In a germanium detector, "a germanium crystal is polarized and the ionization liberated by the electrons can be sensed as an electric pulse of amplitude proportional to the deposited electron energy. If the photon is absorbed, the detector gives a signal proportion to the photon energy" [77].

A basic explanation of how a γ-ray spectrum is constituted is given as follows (see Figure 5.2) [78]: the γ rays produced by a radioactive source interact with the detector in various ways, and produce varying shapes on the γ-ray spectrum that reflect these ways of interacting. Those γ rays that interact via the photoelectric effect will be completely absorbed (by a bound electron), and will thus impart the full amount of their energy to the detector, if the electron stops in the active volume; this will produce a pulse at the full energy of that γ which is called the photopeak. Also, when a photon undergoes Compton scattering followed by a photoelectric effect, all its energy is again deposited, leading to the full-energy peak of the spectrum.

Many Compton-scattering events, however, impart only a portion of their energy to the detector, if the scattered γ ray leaves the detector and only the recoiling electron stops. Thus, Compton events produce a plateau-like area over a wide range of energy values.

Photons with a gamma energy greater than 1.02 MeV can undergo pair pro-

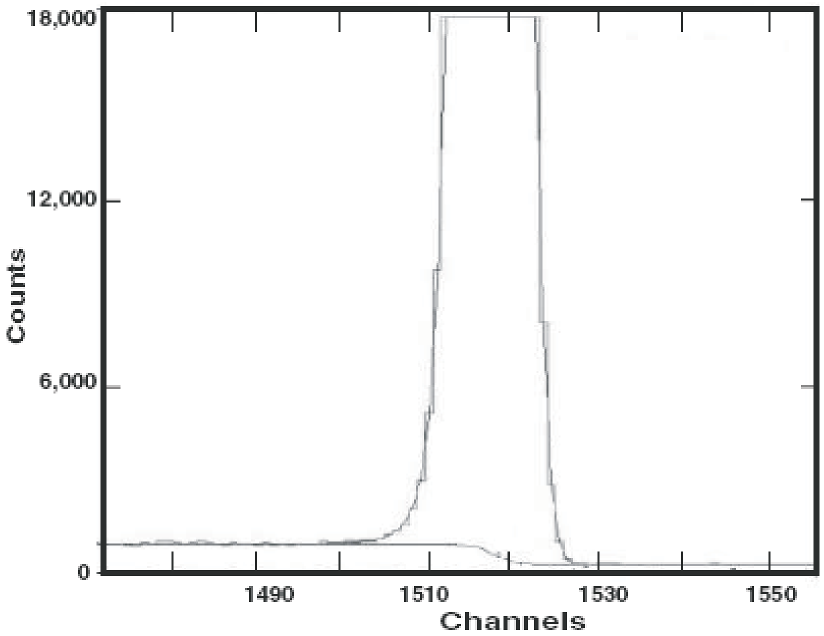

Figure 5.1: Example of a measured 412-keV γ-ray peak together with the fit obtained from GF3. Note that the vertical scale has been greatly expanded to display the low-level background and the quality of the fit to it. The discontinuous line shows the peak, while the smooth line shows the fit to the peak. This spectrum was taken about a week after we began counting. This peak contained a net 1.46 million counts.

duction, in which case the photon will disappear and an electron-positron pair be formed. When this occurs, "The electron and positron then lose their kinetic energy by ionization and the positron then annihilates, generally at rest, with another electron" [77]. "Depending on whether both, one or neither of the annihilation photons is photo-absorbed in the detector, pair production contributes to the full-energy peak or to "single-escape" or "double-escape" peaks" [77]. If present, a "single-escape" peak will be located 511 keV below the full-energy peak, while a "double-escape" peak will be located at twice that distance, or 1,022 keV, below the photopeak [77].

The Compton distribution has an upper limit (or edge) $T_{e(max)}$, whose value can be calculated by the equation:

$$T_{e(max)} = 2E_{\gamma}^{2}/(mc^{2} + 2E_{\gamma}) \tag{5.1}$$

So-called "backscattering" peaks can be produced when photons are reflected off, for example, the sample holder. When this occurs, the backscattering energy can be calculated as:

$$E_{backscatter} = E_{\gamma} - T_{e(max)} \tag{5.2}$$

As an example, for a γ photon of energy 662 keV, after calculation, $T_{e(max)}$ will equal 478 keV; this will be the energy of the Compton edge. The backscattering energy $E_{backscattering}$ will then be 662 keV - 478 keV, or 184 keV. X-ray peaks, typically at low energies, can also be produced by internal conversion from the source.

Regarding the analysis of the temperature-dependence half-life measurements of the nuclide ^{97}Ru, we show a typical γ-ray spectrum in Figure 5.3. Aside from the weak peaks that are due to room background, the only observed γ rays are from the

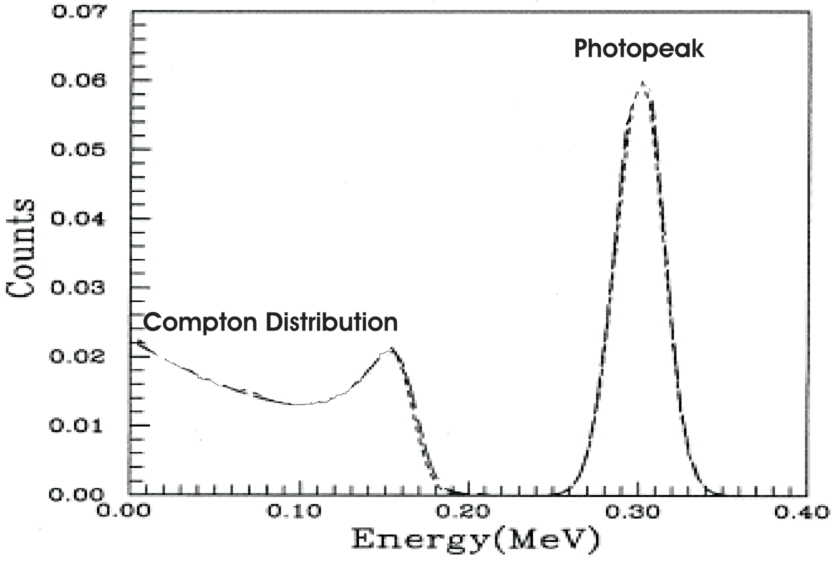

Figure 5.2: Shown here is a basic diagram of the composition of a γ-ray spectrum [78]. The Compton distribution and the photopeak are comprised of various interactions of γ rays from a radioactive source interacting with the detector. See text for a more detailed explanation.

decays of ^{97}Ru ($t_{1/2} = 2.8$ d), ^{103}Ru (39 d), and ^{105}Rh (35 h). The latter nuclide is the daughter of ^{105}Ru (4.4 h), which by the time this spectrum was recorded had already decayed away. The appearance of these three ruthenium isotopes is consistent with their being produced by neutron activation of naturally occurring ruthenium. The 216-keV γ-ray peak from ^{97}Ru is seen to be clear of any other peaks, and to lie on a smooth, though rather high, background.

As with the ^{198}Au measurement, the 216-keV γ-ray peak in each recorded spectrum was analyzed with GF3 [75]. Figure 5.4 shows a sample peak and the fitted background from which its area was determined.

All spectra for ^{97}Ru were subjected to this careful analysis, with the counts recorded in the peaks of interest for each being corrected for residual losses (as was discussed in section IV).

The decay curves were then analyzed by a maximum-likelihood fit, to a single exponential. We used a code based upon ROOT [79], which has previously been tested down to a 0.01% precision using Monte Carlo generated data. The results for both the room temperature and 19 K measurements of ^{198}Au are plotted, as a logarithm against time, in Figures 5.5 and 5.6.

Plotting ln(counts) against time is justified by the following derivation:

$$\Delta N_i = N_i - N_{i+1} \tag{5.3}$$

$$\Delta N_i = N_0 e^{-\lambda t_i} - N_0 e^{-\lambda t_{i+1}} \tag{5.4}$$

$$\Delta N_i = N_0 e^{-\lambda t_i} - N_0 e^{-\lambda (t_i + \Delta t)} \tag{5.5}$$

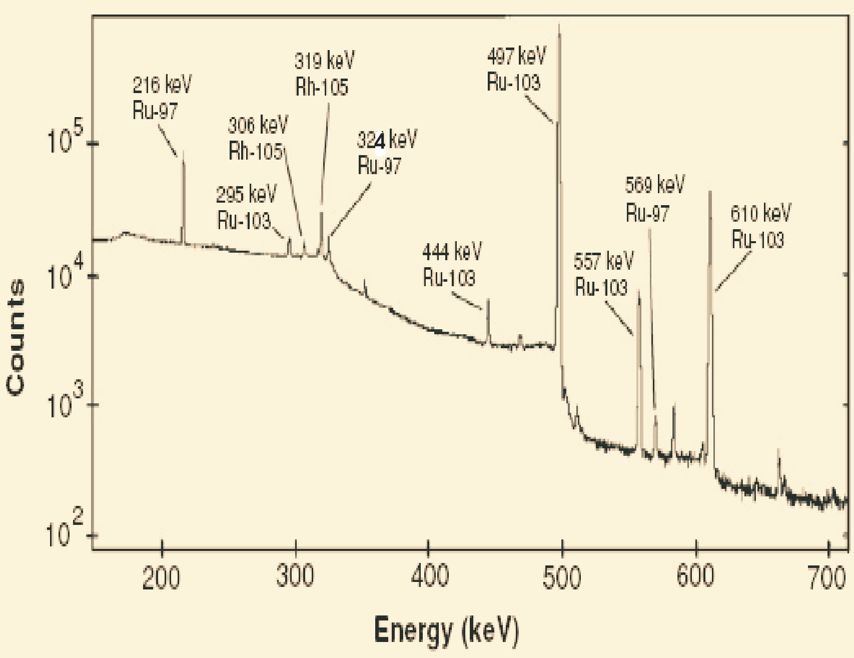

Figure 5.3: The principal peaks from the decays of ^{97}Ru, ^{105}Rh, and ^{103}Ru are marked both by the parent isotope and the energy, in keV. These are all pure peaks with the following two exceptions: the peak at 295 keV consists of approximately 75% ^{103}Ru and 25% ^{226}Ra; and the peak at 610 keV consists of approximately 95% ^{103}Ru and 5% ^{214}Bi. The remaining unmarked peaks are well-known background peaks identified in a separate background measurement.

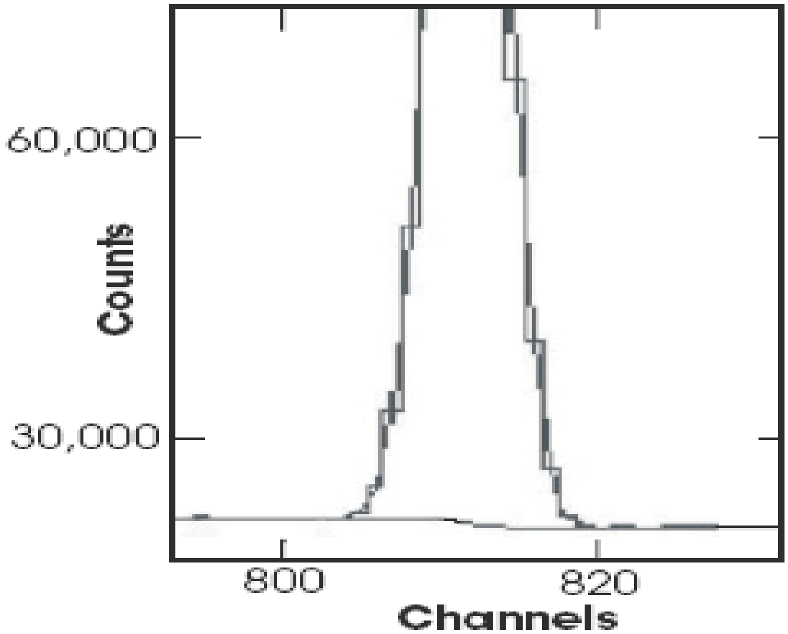

Figure 5.4: Example of a measured 216-keV γ-ray peak together with the fit obtained from GF3. Note that the vertical scale has been greatly expanded to display the low-level background, and the quality of the fit to it. This spectrum was taken about five days after we began counting; the peak contained a net of about 6×10^5 counts.

$$\Delta N_i = N_0 e^{-\lambda t_i}(1 - e^{-\lambda \Delta t}) \tag{5.6}$$

$$ln\Delta N_i = lnN_0 - \lambda t_i + ln(1 - e^{-\lambda \Delta t}) \tag{5.7}$$

$$ln\Delta N_i = -\lambda t_i + C \tag{5.8}$$

In addition to giving the best straight-line fit to the logarithmically-plotted data, the ROOT program also plots, separately, the residuals pertaining to the plot; these residuals are shown at the bottom of Figures 5.5 and 5.6. The residual is the difference, for each data point, between the fit value and the experimental value. For these plots, this difference is then divided by the standard deviation σ to show the relationship between each residual and its corresponding standard deviation.

As another example of a ROOT fit, the result for the gold oxide half-life measurement is shown in Figure 5.7, in which the fitted decay curve is compared with the data in the top panel, and the normalized residuals are shown in the bottom panel.

As a byproduct of our primary measurement on ^{97}Ru we have also extracted from the same spectra half-lives at both temperatures for the isotopes ^{103}Ru and ^{105}Rh, both of which are β^- emitters. For ^{103}Ru, we monitored the 497-keV peak in all 237 spectra; for the shorter lived ^{105}Rh there were only sufficient statistics for us to use 100 spectra, to follow the 319-keV peak (see Fig. 5.3). These peaks were subjected to the same meticulous examination, fitting and analysis as just described for the 216-keV peak of ^{97}Ru.

Now that we have looked at the procedures that were utilized during the analysis of our data. In the next section, we shall discover what the results were for all these

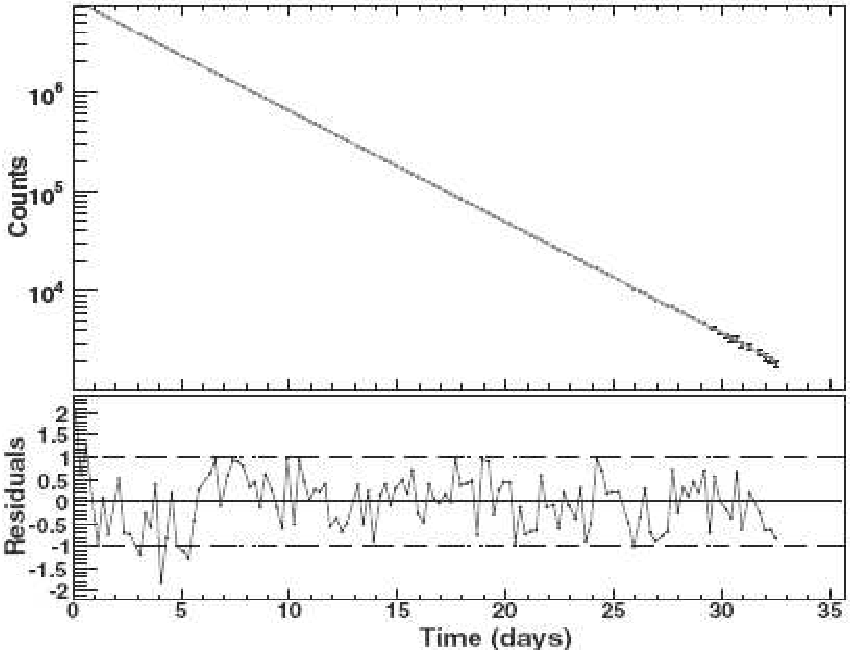

Figure 5.5: The decay of ^{198}Au, in gold at room temperature, as plotted by ROOT. The experimental data appear as dots, with the straight line being a fit to these data. The normalized residuals are shown at the bottom of the figure.

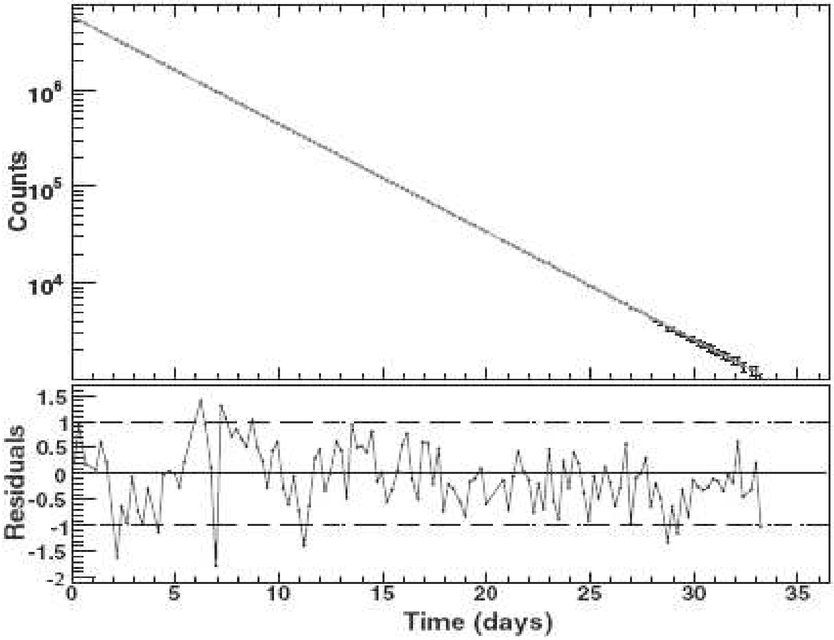

Figure 5.6: The decay of ^{198}Au in gold at 19 K, as plotted by ROOT. The experimental data appear as dots, with the straight line being a fit to these data. The normalized residuals are shown at the bottom of the figure.

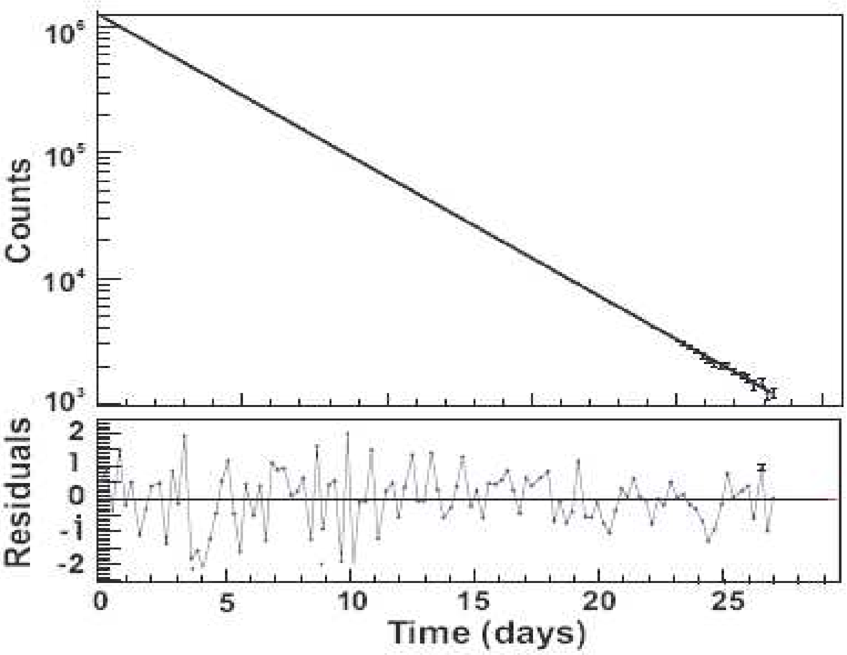

Figure 5.7: The decay of ^{198}Au in gold(III) oxide made at room temperature as an example of a ROOT fit. The experimental data appear as dots in the upper portion of the figure, with the straight line being the fit to these data. The normalized residuals are shown at the bottom.

experiments.

6. RESULTS[1]

6.1 ^{198}Au temperature-dependence

The data in figures 5.5 and 5.6 yielded ^{198}Au half-lives (with statistical uncertainties only) of 2.6949 ± 0.0005 d for the room temperature measurement, and 2.6953 ± 0.0006 d for the measurement taken at 19 K. We can therefore conclude that any difference in half-life between these two temperatures is less than 0.0012 d, or 0.04%. This result thus contradicts the claim [4] that there is a 90 times larger difference, 3.6 ± 1.0%, between the half-lives when measured at essentially the same two temperatures.

The half-life values taken from these computer fits do not yet include the uncertainty in the residual loss corrections, since they are correlated for the two measurements, and therefore do not contribute to the difference between them. Before our

measurements can be compared with previous precision measurements of the ^{198}Au half-life, however, this systematic uncertainty is incorporated, and gives the resulting values of 2.6949 ± 0.0008 d and 2.6953 ± 0.0009 d for the room temperature and 19 K measurements, respectively.

Both these results agree well with previous measurements which have been made

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[1] Reprinted with permissions from "The half-life of ^{198}Au: High-precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob and J. C. Hardy, 2007. The European Physical Journal A, **34**, 271-274, Copyright 2007 by European Physical Journal; "Half-life of the electron-capture decay of ^{97}Ru : Precision measurement shows no temperature dependence" by J. R. Goodwin, V. V. Golovko, V. E. Iacob, and J. C. Hardy, 2009. Physical Review C, **80**, 044501, Copyright 2009 by Physical Review C; "Measurement of the half-life of ^{198}Au in a non-metal: High-precision measurement shows no host-material dependence" by J. R. Goodwin, N. Nica, V. E. Iacob, A. Dibidad and J. C. Hardy, 2010. Physical Review C, **82**, 044320, Copyright 2010 by Physical Review C; and "Do radioactive half-lives vary with the Earth-to-Sun distance?" by J. C. Hardy, J. R. Goodwin and V. E. Iacob, 2012. Applied Radiation and Isotopes, **70**, 1931-1933, Copyright 2012 by Applied Radiation and Isotopes.

on ^{198}Au at room temperature. In table 6.1 and figure 6.1, we compare our results to those obtained since 1965 that have 0.15% precision, or better. (The room temperature result of Spillane *et al.* has 0.7% precision and was not included in the figure because its uncertainty would have been off scale.) It is evident that these previous measurements do not agree particularly well even with one another, at the precision claimed by the original authors: the normalized χ^2 for their average is 8. Although the uncertainty that we associate with this average in the table has already been inflated by the square root of the normalized χ^2, it could be argued that this still does not accurately reflect the actual uncertainty of the average. Regardless, our half-life results at both temperatures are in total agreement with it.

After we published our result [15] it was confirmed by two other new measurements of ^{198}Au, conducted both at room temperature and at very low temperature, which set limits for half-life change of 0.13% in a Al-Au alloy host [80] and of 0.03% in gold metal [81].

6.2 ^{97}Ru temperature dependence

A total of 229 spectra were evaluated by the careful analysis (see Section V), with the counts which were recorded in the 216-keV peak for each being corrected for residual losses (as discussed in Section IV). The results for the room temperature as well as the 19 K measurements are plotted, as a function of time, in Figs. 6.2 and 6.3. The decay curves were then analyzed as described above for ^{198}Au.

The data in Figs. 6.2 and 6.3 yield ^{97}Ru half-lives (with statistical uncertainties only) of 2.8370(13) d for the room temperature measurement, and 2.8382(13) d for the measurement conducted at 19 K. The difference between two results is 0.0012(18) d, which gives an upper limit of 0.0030 d, or 0.1%, on any temperature-dependent difference, at the 68% confidence level.

Table 6.1: Measurements of the ^{198}Au half-life made since 1965 (at room temperature).

Half Life (d)	Reference	Year
2.694(4)	Anspach *et al.* [82]	1965
2.695(2)	Vuorinen and Kaloinen [83]	1969
2.696(4)	Costa Paiva and Martinho [84]	1970
2.6946(10)	Cabell and Wilkins [85]	1970
2.693(3)	Debertin [86]	1971
2.695(3)	Goodier *et al.* [87]	1971
2.6937(2)	Merritt and Gibson [88]	1977
2.6935(4)	Rutledge *et al.* [89]	1980
2.695(2)	Hoppes *et al.* [90]	1982
2.6966(7)	Abzouzi *et al.* [91]	1990
2.69517(21)	Unterweger *et al.* [92]	1992
2.69573(14)	Unterweger and Lindstrom [93]	2004
2.6924(11)	Lindstrom *et al.* [94]	2005
2.69500(27)	Weighted Average	
This work:		
2.6949(8)	Room Temperature	2007
2.6953(9)	19 K	2007

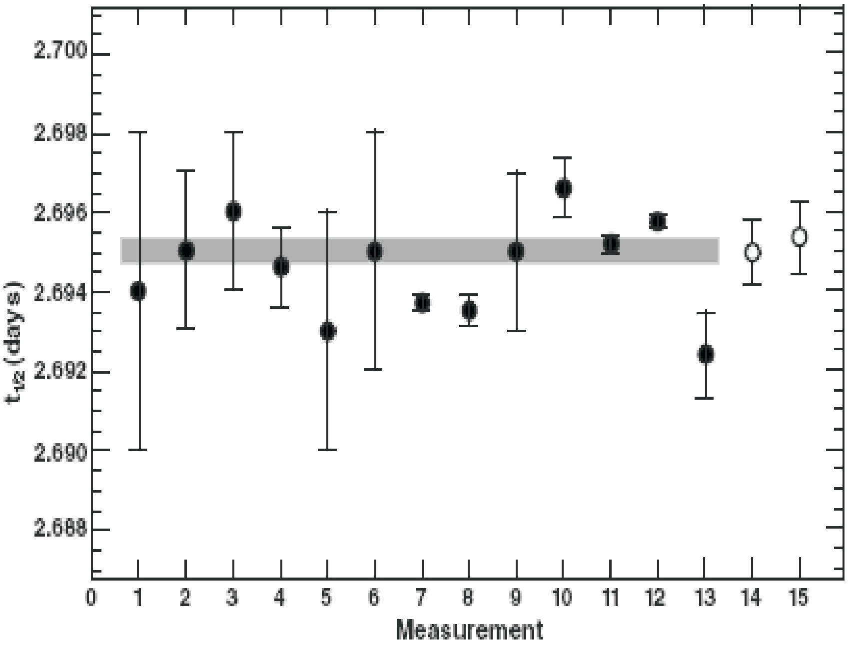

Figure 6.1: The solid circles indicate the precise measurements of the ^{198}Au half-life that have been made since 1965; they are plotted in chronological order, from left to right. The gray band shows the weighted average, with the uncertainty, of these previous measurements. The open circles give the results of the present measurement, with the 19 K measurement to the far right. The data in the figure are those tabulated in table 6.1.

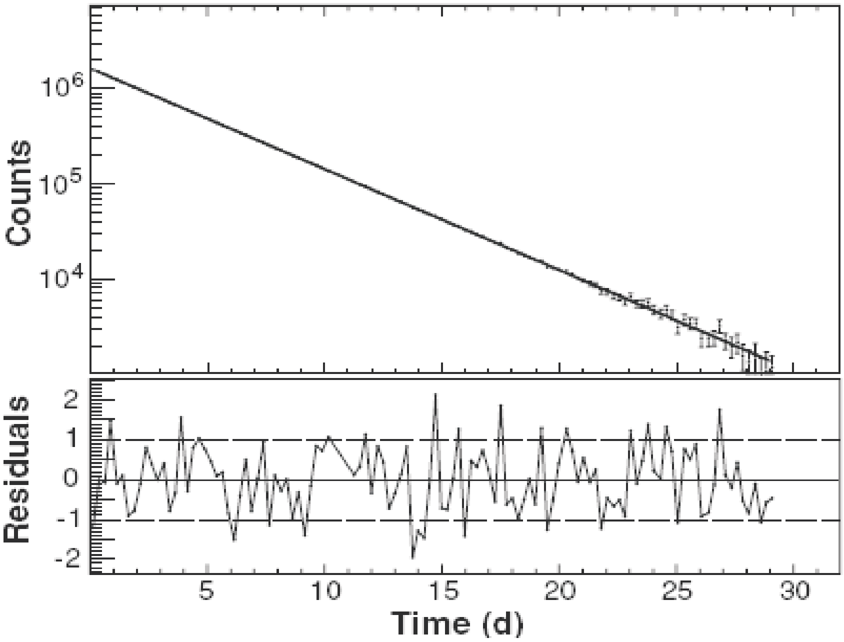

Figure 6.2: The decay of ^{97}Ru in ruthenium metal, at room temperature. The experimental data appear as dots, with the straight line being the fit to these data. The normalized residuals are shown at the bottom of the figure. The dashed lines in the residuals plot represent ±1 standard deviation from the fitted value.

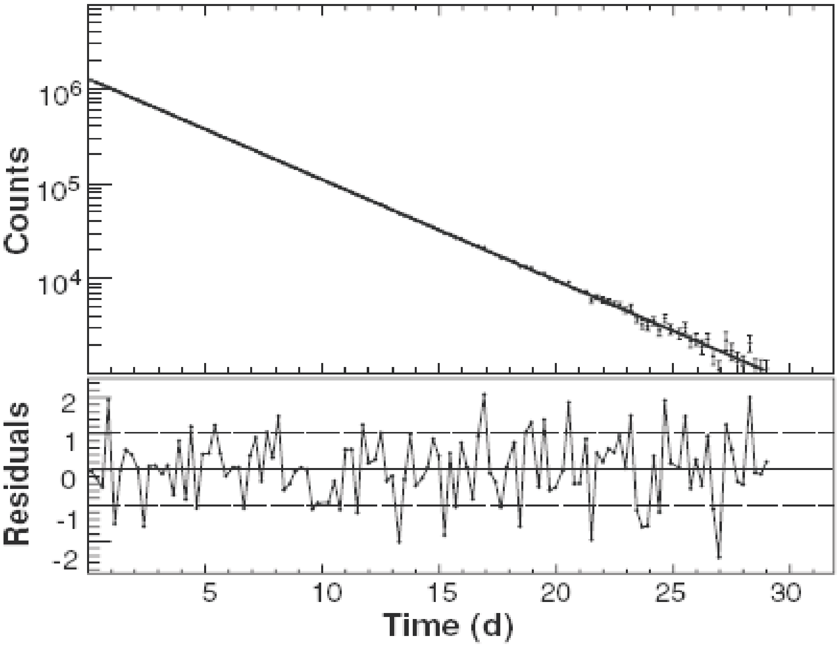

Figure 6.3: The decay of ^{97}Ru in ruthenium metal, at 19 K. The experimental data appear as dots, with the straight line being the fit to these data. The normalized residuals are shown at the bottom of the figure. The dashed lines in the residuals plot represent ±1 standard deviation from the fitted value.

For our ^{97}Ru measurements to be compared with previous measurements of the ^{97}Ru half-life, the systematic uncertainty as described for ^{198}Au previously, should now be incorporated; and when this is done the results are 2.8370(14) d and 2.8382(14) d for the room temperature and 19 K measurements, respectively. These values are then compared with previous measurements of the ^{97}Ru half-life in Table 6.2 and Fig. 6.4; in these it can be seen that our results at both temperatures are much more precise than, but are entirely consistent with, the previous ones, all of which were presumably made at room temperature.

For the isotopes ^{103}Ru and ^{105}Rh, incorporating only statistical uncertainties, we obtained half-life values (for ^{103}Ru) of 39.210(16) d at room temperature, and 39.219(25) d at 19 K; these are statistically the same within 0.1%. For ^{105}Rh, our half-life values (with statistical uncertainties only) are 35.357(36) h at room temperature and 35.319(23) h at 19 K, again the same, but in this case within 0.2%.

As in making the temperature comparison with ^{97}Ru, we have to this point quoted half-life values for ^{103}Ru and ^{105}Rh that do not yet include the (correlated) uncertainty, attributable to residual losses (see Section IV). We include that at this point so that we can compare our results with previous half-life measurements. Our final half-life results for ^{103}Ru then become 39.210(38) d at room temperature, and 39.219(35) d at 19 K; and for ^{105}Rh our results are 35.357(37) h at room temperature and 35.310(24) h at 19 K. It is worth noting that the effect of the residual losses on the uncertainty of the ^{103}Ru half-life is much greater than it is for the ^{105}Rh half-life. Since ^{103}Ru is much longer lived, our data only encompass a little more than one half-life for this nuclide, and during that time the overall count rate in our detector has decreased significantly.

Unlike the situation for the other two radionuclides studied in the ruthenium series of measurements, the half-life of ^{103}Ru has been measured rather precisely in

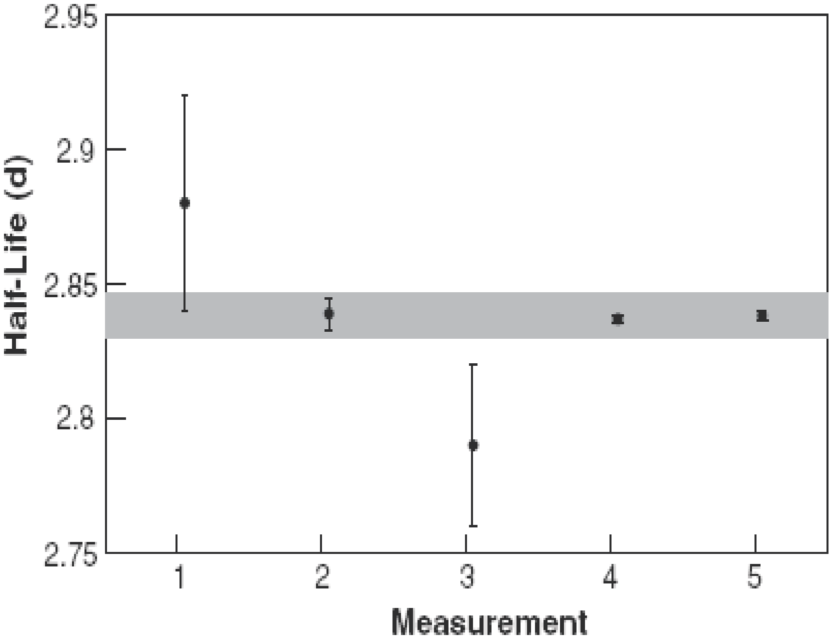

Figure 6.4: The data points represent all of the published measurements of the ^{97}Ru half-life that have been quoted with better than 2% precision. The results are plotted in chronological order from left to right, measurements 1-3 being those of Katcoff *et al.* [97], Silvester *et al.* [99], and Kobayashi *et al.* [100]. The shaded area represents the weighted average of these measurements. Measurements 4 and 5 are the room-temperature and 19 K results of the present measurement. The data in the figure are taken from those tabulated in Table 6.2.

the past, with four of the previous results being of comparable precision to our current ones. Unfortunately however the earlier results are not particularly consistent with one another; this can be seen in Table 6.3. The normalized χ^2 for the average of all previous measurements is 3.0, which results in our needing to scale up the uncertainty assigned to that average by a factor of 1.7. In comparison with this average value, our results are a bit low, although the discrepancey is not very significant statistically. Note also that our results are completely consistent with the 1981 value obtained by Miyahara *et al.* [101].

For the ^{105}Rh half-life, only three previous measurements exist, none of which is more recent than 1967. They are listed in Table 6.4. Strikingly, the earliest measurement [102] has the tightest, ±0.06%, uncertainty and a half-life value that disagrees completely with the two later measurements. The weighted average of all three measurements yields a normalized χ^2 of 22 and, as shown in Table 6.4, its uncertainty consequently again requires scaling, this time by a factor of 4.7. Under the circumstances, it seems more reasonable not to use this average value, but simply to disregard the offending measurement and average the two remaining, mutually consistent, results [103, 104]. When compared with this new average, our results are a factor of two more precise and lie slightly lower. Considering that even the two previous measurements that have been retained are more than 40 years old and that the difference between their average and our recent results is less than two standard deviations, there seems little reason for concern.

6.3 Au_2O_3 host-material dependence

After performing the careful analysis of each spectrum as described in section IV, we then analysed the decay curve in the same manner as for the ^{198}Au and the ^{97}Ru measurements above. The resulting fit for the gold oxide half-life measurement is

Table 6.2: Measurements of the ^{97}Ru half-life made since 1946.

Half Life (d)	Reference	Year
2.8(3)	Sullivan *et al.* [95]	1946
2.8(1)	Mock *et al.* [96]	1948
2.88(4)	Katcoff *et al.*[97]	1958
2.9(1)	Cretzu *et al.* [98]	1966
2.839(6)	Silvester *et al.* [99]	1979
2.79(3)	Kobayashi *et al.* [100]	1998
2.838(6)	Weighted Average	
This work:		
2.8370(14)	Room Temperature	2009
2.8382(14)	19 K	2009
0.0012(18)	Difference	

Table 6.3: Measurements of the ^{103}Ru half-life quoted with sub-percent precision

Half Life (d)	Reference	Year
39.5(3)	Flynn *et al.* [105]	1965
39.35(5)	Debertin [106]	1971
39.254(8)	Houtermans *et al.* [107]	1980
39.214(13)	Miyahara *et al.* [101]	1981
39.260(20)	Vaninbroukx *et al.* [108]	1981
39.272(16)	Walz *et al.* [109]	1983
39.250(10)	Weighted Average (Scale Factor 1.7)	
This work:		
39.210(38)	Room Temperature	2009
39.219(35)	19 K	2009
0.0009(30)	Difference	

given in Figure 6.5, in which the fitted decay curve is compared with the data in the top panel and the normalized residuals are shown in the bottom panel. The ^{198}Au half-life obtained from this fit (with statistical uncertainty only) is 2.6948(9) d. The corresponding normalized χ^2 is 0.74, which gives a confidence level of 99%.

The corresponding room-temperature value for the ^{198}Au half-life, as measured in our pure-gold host material, was reported by us [15] to be 2.6949(5) d. The difference between these two results is 0.0001(10) d or 0.004(38)%. Both measurements were made under the same conditions and the data from both have been corrected for residual losses; however, the uncertainty in that correction has not yet been applied because it is correlated for the two measurements and does not contribute to the difference between them.

For the present measurement the systematic uncertainty associated with the residual-loss correction is small when compared to the statistical uncertainty; this results in the total uncertainty being unchanged from the statistical uncertainty. So, our final result is thus 2.6948(9) d. This value is in excellent agreement with 2.695 00(27) d, the weighted average of all previous measurements of the gold half-life (see table 6.1).

6.4 Earth-Sun distance-dependence

Seven measurements were performed with regard to the Earth-Sun study; the first three of these have already been described. Following our decision to pursue a study of a possible Earth-Sun distance dependence on the ^{198}Au half-life, four more ^{198}Au half-life measurements were performed. The results for all seven measurements are given in Table 6.5. The uncertainties quoted there are purely statistical, and do not include a provision for the uncertainty in the correction for residual losses. This is because that correction is correlated for all seven measurements, and does not

Table 6.4: Measurements of the ^{105}Rh half-life.

Half Life (h)	Reference	Year
35.88(2)	Brandhorst and Cobble [102]	1962
35.4(1)	Pierson [103]	1965
35.47(8)	Kobayashi [104]	1967
35.84(9)	Weighted Average (Scale Factor 4.7)	
35.44(6)	Weighted Average of [103] and [104]	
This work:		
35.357(37)	Room Temperature	2009
35.319(24)	19 K	2009
0.038(43)	Difference	

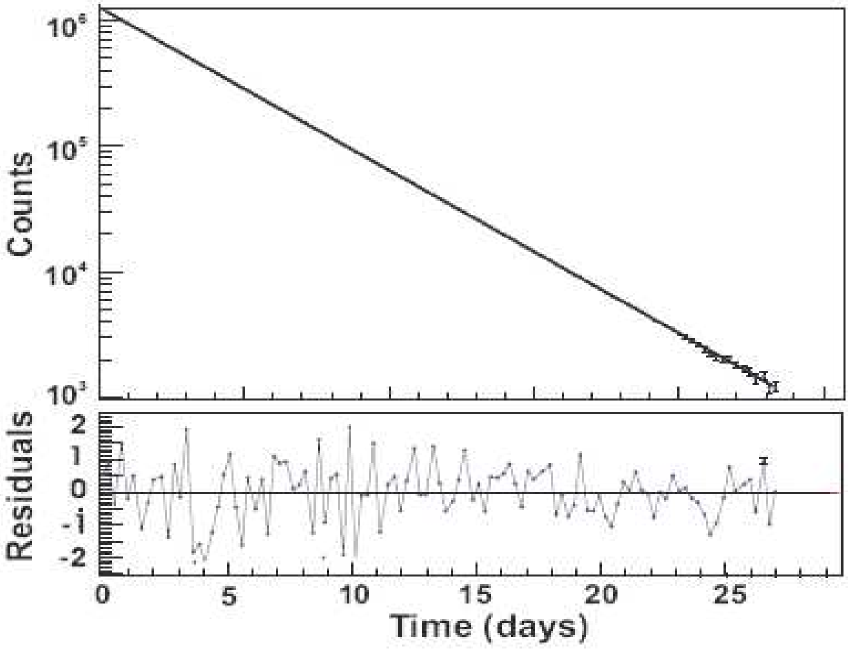

Figure 6.5: The decay of ^{198}Au in gold(III) oxide made at room temperature. The experimental data appear as dots in the upper portion of the figure, with the straight line being the fit to these data. The normalized residuals are shown at the bottom.

contribute to the differences among them.

In Table 6.5, each measurement is characterized by a date and a time, which is taken to be one mean-life of ^{198}Au ($\tau = t_{1/2}/\ln 2 = 3.89$ d) after the actual start time of the measurement. The table also shows how the time of each measurement relates to the time of the preceding perihelion of the Earth-Sun system. Clearly there is no dependence of the half-life on this parameter within our limits of uncertainty. In fact, the weighted average of the seven measured half-lives is 2.69445(20) d with a normalized χ^2 of 0.74 and a confidence level of 64%. Since the BNL measurements were of activity, not half-life, we show our half-life results for ^{198}Au expressed as decay rates, λ (= $\ln 2/t_{1/2}$), in the final column of Table 6.5.

In Figure 6.6 we show the ROOT fit for our fifth dedicated Earth-Sun measurement. The experimental data appear as dots in the figure, and the straight line fits the data points.

Before we continue the discussion of our results, let us review briefly the Brookhaven study. This study, conducted between 1982 and 1986, involved repeatedly measuring the activities of ^{32}Si ($t_{1/2}$ = 172 y), and ^{36}Cl (a long-lived isotope with $t_{1/2}$ = 1.6 x 10^3 y) directly, and then obtaining the ratios of ^{32}Si to ^{36}Cl, over a period of four years. These ^{32}Si to ^{36}Cl ratios were then converted into a function $U(t)\equiv[\dot{N}(t)/\dot{N}(0)]\exp(+\lambda(t))$, which was then normalized and plotted as a function of time (here $\dot{N}\equiv dN/dt$). A graph of the value of $1/R^2$, R being the mean Earth-Sun distance (149,597,870.7 km) as given in astronomical units (AU), was then superimposed upon the plot.

How do our results compare with the BNL-observed activity oscillations, upon which Jenkins, Fischbach *et al.* base their claims? In Figure 6.7 we present the BNL activity results [60] as displayed by Jenkins *et al.* [59]. The activity ratios ^{32}Si/^{36}Cl are normalized to their average and appear as gray circles with error bars, which are

Table 6.5: The dates of, and the results from, our seven measurements of the half-life of ^{198}Au. The date in each case is taken to be the starting time of our measurement plus one mean-life of ^{198}Au (3.89 d). The decay rate, λ, is related to the half-life, $t_{1/2}$, by $\lambda = \ln 2/t_{1/2}$. The column entitled "Perih." gives the number of days that have passed since the preceeding perihelion.

No.	Date (y/m/d/h)	Perih.	$t_{1/2}$ (d)	λ (d^{-1})
1	2007/06/24/1.2	171.2	2.6953(6)	0.25717(6)
2	2007/08/13/19.2	222.0	2.6949(5)	0.25721(5)
3	2009/06/14/21.2	161.3	2.6948(9)	0.25722(9)
4	2009/09/25/19.9	264.2	2.6944(5)	0.25725(5)
5	2009/11/08/17.6	308.1	2.6942(4)	0.25727(4)
6	2010/01/10/17.2	7.7	2.6940(5)	0.25729(5)
7	2010/03/07/16.6	63.7	2.6942(5)	0.25727(5)

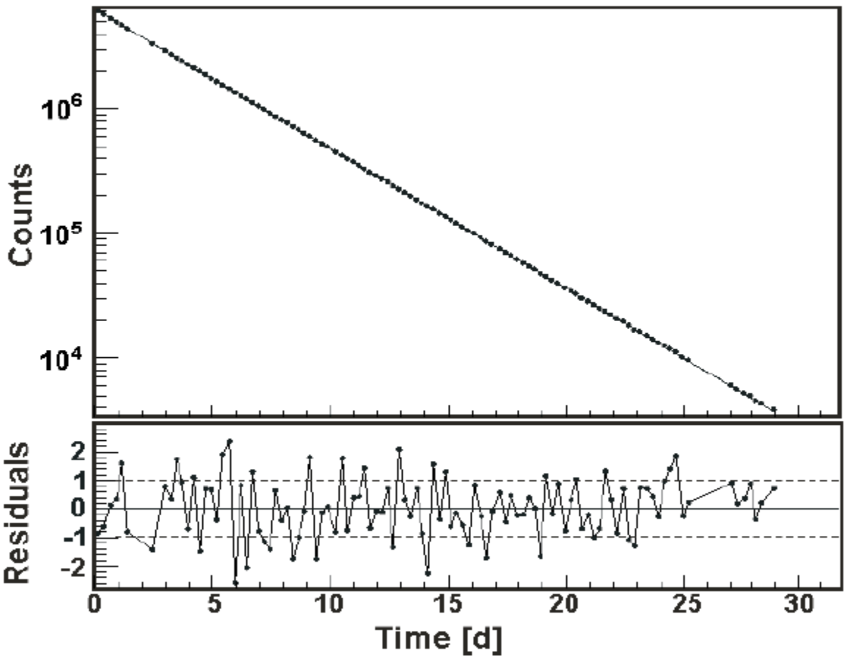

Figure 6.6: The decay of ^{198}Au as obtained in our dedicated Earth-Sun measurement #5. The experimental data appear as dots, with error bars; the straight line is a fit to these data. The normalized residuals are shown at the bottom of the figure. The dashed lines in the plot of residuals represent ±1 standard deviation from the fitted value.

then plotted against the dates of their measurement over a period of four years from early 1982 to early 1986. The dotted curve shows the $1/\mathrm{R}^2$ behavior of the Earth-Sun distance over the same period, which is very nearly a pure sine wave. We have therefore fitted the BNL data with a sine wave, which has a fixed one-year period, but a variable amplitude and phase shift. A least squares fit to the data, shown as the dashed curve in the figure, has an amplitude of $7.9(3) \times 10^{-4}$ and a phase shift of 35(2) d relative to the $1/\mathrm{R}^2$ plot. This phase shift was also noted by Jenkins *et al.* [59].

The time scale for our measurements has been displaced by exactly 25 years from the BNL scale, so our data appear with the same synchronization of perihelion. The horizontal shaded band shows the uncertainty limits of one-standard-deviation on the average value from our results. As we have already noted, our data are statistically consistent with a constant half-life value to within a relative precision of $\pm 7 \times 10^{-5}$, an order of magnitude smaller than the amplitude of the oscillations attributed to the BNL data.

It is also interesting to compare our results for the ^{198}Au half-life with previously published values for the same quantity. However, the uncertainties associated with $t_{1/2}$ in Table 6.5 are purely statistical. Although the correction for residual losses has been incorporated in the half-life value itself, the uncertainty in the correction, being common to all, has not been applied to the uncertainties in the table. If we now apply that common uncertainty to the average value of the ^{198}Au half-life, we obtain the final result:

$$t_{1/2}(^{198}Au) = 2.69445 \pm 20(stat.) \pm 25(syst.)d. \tag{6.1}$$

This value agrees completely with 2.6950(3) d, which is the weighted average

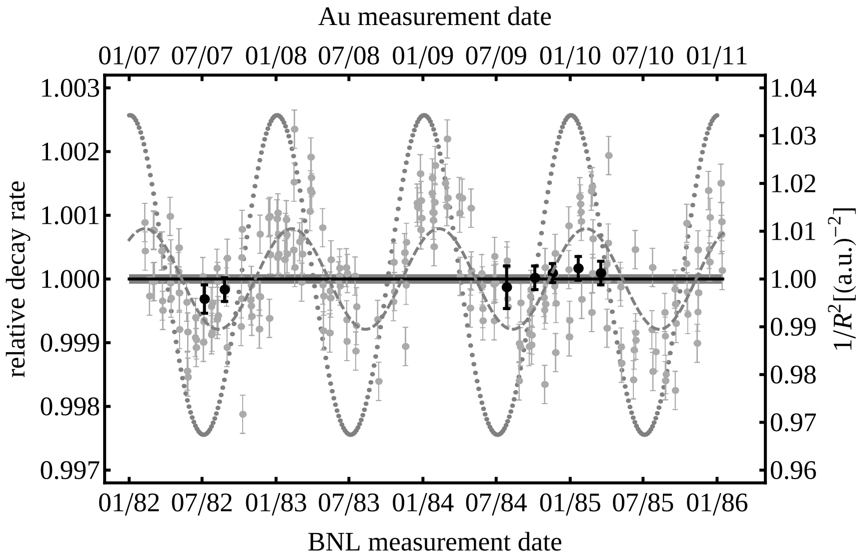

Figure 6.7: Here, the BNL data for the activity ratio ^{32}Si/^{36}Cl as measured by Alburger *et al.*[60] as published by Jenkins [59], are plotted as gray circles with error bars (referred to the vertical axis at the left) against the dates of their measurement between 1982 and 1985 (horizontal axis at the bottom). The dotted curve shows the $1/\mathrm{R}^2$ behavior of the Earth-Sun distance, where R is measured in astronomical units, a.u. (vertical scale at the right), over the same period; and the dashed curve gives our fit to the BNL data (see text). Our seven results for the decay rate of ^{198}Au normalized to their average value (with the same vertical scale as the BNL data) are plotted as black circles with error bars against their dates (shown on the horizontal axis at the top, which is shifted exactly 25 years as compared to the bottom scale).

of all thirteen previously published measurements that claimed better than 0.15% precision. It also agrees with, but is more precise than, 2.6950(7) d, the value recommended in the most recent IAEA data evaluation (IAEA, 2007).

It is interesting to note at this point that, of the numerous articles that have been published on the subject of half-lives being influenced by the Earth-Sun distance recently, only two publications have put forth experimental results contradicting temporal oscillations in the radioactive decay rates. One of these is by Cooper [110], who utilizes data taken from the Cassini spacecraft. Cooper, of the Fermi National Accelerator Laboratory, has attempted to find a correlation between the Earth-Sun distance and the decay of ^{238}Pu ($t_{1/2}$ = 87.7 y). From data Cooper obtained from the NASA Jet Propulsion Laboratory, he has plotted the Earth-Sun distance superimposed over a plot of the electrical power output of the RTG (Radioisotope Thermoelectric Generators) aboard the Cassini Spacecraft, which was launched in 1997. The Cassini spacecraft is powered by three of these RTG's; each RTG represents a large ^{238}Pu source. ^{238}Pu decays by α-decay with a half-life of 87.7 y. Cooper has found "no significant deviations from exponential decay over a range of 0.7 - 1.6 A.U. (one A.U., or astronomical unit, represents the mean distance between the Earth and the Sun. One A.U. = 149,598,000 meters). The Cassini RTG power data exclude any variation of the ^{238}Pu nuclear decay rate correlated with the distance of the source from the sun to a level 350x smaller than the effect reported by Jenkins *et al.*". And in the second article, Norman *et al.* [111] find no evidence of correlations between decay rates and the Earth-Sun distance, after having analyzed data which had previously been published regarding long-lived α, β^+, β^- and EC emitters.

Also, we mention here that, as regards the oscillatory behavior of the decay rate observed in the PTB data, Schrader [112] has recently pointed out that the variations in the results from the PTB group vanish, or at least change their structure totally, when a different technique is used for current measurement.

7. CONCLUSIONS

7.1 ^{198}Au

One of the measurements that was reported in the Rolfs research group's series was performed by Spillane *et al.* [4], as mentioned above, on gold metal, in which the increase in half-life was 3.6 ± 1.0 %. We repeated their experiment, and recorded the decay with much better statistics, for over ten half-lives; this applied to both the room temperature and the 19 K measurements. We were able to obtain 0.04% precision, an improvement of a factor of 25 over the precision acquired by Spillane *et al.*. We did not find a temperature-dependent β^--decay half-life change, for ^{198}Au in a metallic gold environment. We have demonstrated that any difference in half-life between room temperature and 19 K must be less than 0.04 %, a factor of 90 lower than the difference claimed by Spillane *et al.*. Our half-life results agree well with the average of previous precision room temperature measurements of the ^{198}Au half-life. The limit that we obtained for any possible difference was a factor of 500 less than the predictions of the "Debye plasma" theory. This effectively rules out any significant screening from a "Debye plasma" in the case of β^--decay, at least for ^{198}Au.

7.2 ^{97}Ru

We now turn to our conclusions regarding the possibility of half-life change in a nuclide that decays via EC, ^{97}Ru. We have measured the half-life of ^{97}Ru, in ruthenium metal, at room temperature and at 19 K, and have found the results to be the same within 0.1%.

Six years ago, Wang *et al.* [5] reported half-life measurements of another pure electron-capture emitter, ^{7}Be, sited in both palladium and indium metals, in which they observed differences of 0.9(2)% and 0.7(2)%, respectively, between room temperature and 12 K. In that context, their result for ^{7}Be was understood to be the indication of a generic property of all EC decays rather than a unique property of ^{7}Be. Because of its unique atomic properties, however, ^{7}Be is well-known to exhibit half-life changes in response to changes in chemical combination, or to being placed within Bucky balls, etc. ^{7}Be is clearly unique in that a substantial portion of its decays involve valence electrons. So, the finding of a half-life change by Wang for ^{7}Be is in itself not surprising. It won't do, however, to then attempt to make that finding apply to all nuclides that decay by EC.

While we obviously cannot comment on the validity of the ^{7}Be measurement itself, we can certainly refute any suggestion that the half-lives of EC decays - in general - exhibit significant temperature dependence, when the source is placed in a metal host. Wang *et al.* [5] used their model to calculate that the half-life of ^{7}Be in a metal should change by 1.1% between $T = 293$ and 12 K, a result that agrees reasonably well with their measured values.

Using the same model, we calculated that the half-life change for the ^{97}Ru decay used in our case should be 11.2% between $T = 293$ and 12 K and 8.4% between T $= 293$ and 19 K, the temperature we obtained. In our study, as for our previous study of ^{198}Au, we found no temperature dependence in the results. We were able to set an upper limit of 0.1% on any half-life change over this temperature range, nearly two orders of magnitude less than this model prediction. Our upper limit is also an order of magnitude below the effect which had been claimed for the isotope ^{7}Be [5]. We had previously demonstrated that the Debye model has no validity for β^- decay [15]; we could now state with equal confidence that it also does not apply to EC decay [16].

As a byproduct of this primary measurement, we also obtained half-life data for two more β^- emitters, ^{103}Ru and ^{105}Rh, at room temperature and 19 K. These results, though slightly less precise than our measurements on the β^- decay of ^{198}Au [15], nevertheless confirm our previous conclusion for that decay mode. With any temperature dependence for β^+ decay also now ruled out at the 0.04% level [81], it has become clear that there is no reason to doubt the accuracy of nuclear weak-decay half-lives that have been quoted over the past decades with subpercent precision; and without accounting for the host material or temperature. As has always been believed, those parameters indeed do not affect the result, at least not above the 0.1% level. Also, in all three cases, ^{97}Ru, ^{103}Ru, and ^{105}Rh, our measured half-lives are consistent with, and in two cases are substantially more precise than, previous measurements.

Also regarding electron-capture decay, a new measurement of the ^{154}Eu decay, in copper [80], uncovered no temperature dependence greater than 0.3%, but another measurement [52] did actually find a small change in the ^{7}Be half-life - 0.22(8) % - depending on whether the host material was a conductor (Cu or Al) or an insulator (Al_2O_3 or PVC), with both measurements being at room temperature. In neither case, however, was the result as precise as that which had been achieved for β^- and β^+ decays. Since ^{7}Be is well-known to show effects on half-life from its chemical environment, it is difficult to know with certainty the cause of any observed effect, and impossible to generalize its behavior to the electron-capture decay of other nuclei with the K-shell electrons being shielded more fully from the external environment. In this case, one can certainly speculate that changes in half-life could occur by changing the electron density near the nucleus via chemical combination.

Although we did not perform an experiment to study the possible temperature-dependence in β^+ ourselves, Ruprecht *et al.* have done so [81]. This study placed an upper limit on the temperature dependence of the β^+-decay of ^{22}Na at 0.04%, which was nearly two orders of magnitude below the claim in the case made by Limata *et al.*.

Also, for α decay, the ^{210}Po measurement has not yet been repeated, but low-temperature measurements on a variety of other α emitters [113], [114] have set upper limits for this type of decay of 1% on any possible temperature dependence for those cases. Although this 1% limit is significantly smaller than the temperature dependence which was claimed to have been observed in Ref. [3], it is considerably less stringent than the limits which were obtained for the β^- and β^+ decays mentioned above.

7.3 Au_2O_3

We have measured the half-life of ^{198}Au in gold(III) oxide at room temperature. This result, obtained with the decaying nuclei sited in this insulating medium, is consistent with a half-life result we published previously for ^{198}Au sited in pure gold, a conductor. We established that the difference between the half-lives measured in an insulator and in a conductor is less than 0.04%, with a confidence level of 68% (one standard deviation). This limit is more than two orders of magnitude lower than the 7% difference predicted by the "Debye plasma model" [4].

7.4 Earth-Sun distance

We have measured the half-life of ^{198}Au on seven different occasions, which occasions spanned the full range of Earth-Sun distances, from perihelion to aphelion. Each measurement covered ten half-lives of the ^{198}Au decay, and each measurement established the half-life with high precision. The results were consistent with one another to a relative precision of $\pm 7 \times 10^{-5}$, in contradiction to previous claims (*e.g.* [59]) that half-lives changed by as much as $\pm 7.9 \times 10^{-4}$ over each cycle of the Earth-Sun distance. Our average half-life result also agrees well with the average of (extensive) world data.

Because we have depended upon direct half-life measurements of a nuclide of relatively short half-life, rather than upon separate activity measurements which were spaced over a long period of time, we have been able to substantially lower the potential effects of environmental and instrumental variations. In the performance of these measurements, we have observed no systematic oscillations in the half-life of ^{198}Au. We can also set an upper limit on their amplitude, of $\pm 0.02\%$.

The idea of Ockham's razor - that is, the concept that of several competing theories, the simplest one is to be preferred - could be invoked at this point to argue that we need go no farther than to demonstrate that a simple constant fits our data. That is, we could simply claim that since the data can be fitted with a straight line, there is no need for further analysis.

However, for the sake of argument (i.e. playing Devil's advocate) we have taken another step, and have performed a least-squares fit to our data - just to see how such a fit would compare with Jenkins' claims. We did this by applying the same sine wave - as was mentioned previously in section 2, with a one-year period - to our data, as we used when fitting the BNL data. When we fix the phase shift for this sine wave at 35 d, the value obtained from the BNL data, we obtain an amplitude of 1.7(5) $\times 10^{-4}$, with a normalized χ^2 of 0.22. If we then choose to let both the amplitude and the phase shift vary, the amplitude remains about the same, but the phase shift goes to zero and the normalized χ^2 becomes an unreasonable 0.07. Clearly there is no statistically significant support for the interpretation that Jenkins, Fischbach *et al.* have applied to the BNL data. Our results demonstrate that the ^{198}Au half-life is independent of the Earth-Sun distance to within $\pm 7 \times 10^{-3}$ % and, even if one postulates the possibility of a half-life dependence on that distance, the upper limit on its amplitude is 2×10^{-2} %. In either case, these limits are well below the 8×10^{-2} % number obtained from the BNL data on ^{32}Si. Whatever caused the oscillation in the BNL data, it cannot have been principally due to oscillations in the Earth-Sun distance.

7.5 General conclusions

Our concern in undertaking these measurements was for the integrity of precise half-lives measured in the past. Because physical conditions were believed to have no influence on half-lives, no care was taken in the past to select a particular host material or even to specify the temperature at which a measurement was made. Our main concern was with the half-lives of superallowed $0^+ \rightarrow 0^+$ β^+ emitters, which are essential to fundamental tests of the standard model [7]. Their precision has typically been quoted to less than 0.05%, well below the temperature and host-material dependence claimed by the measurements in Refs. [3, 4, 5].

Our results, together with previous measurements of ours [15, 16, 17] and others [81], effectively refute all the predictions of the Debye plasma model as they apply to β^-, β^+, and electron-capture decays, and also contradicts the measurements that initially supported those predictions [3, 4, 5].We can now state with confidence that, at the level of 0.05%, half-lives are affected neither by temperature changes between 19 K and 295 K nor by the resistivity of the host medium in which they are located. *There is no need to revisit past measurements of half-lives quoted to high precision.*

So it seems that as we finish our discussion of the possibility of external factors producing changes in radioactive half-life, we return again to that point whence we began. As you will remember, we started this thesis by relating the historical belief of the immutability of radioactive half-lives, regardless of changes in temperature, external pressure, host material (or as we can now add, the distance between the Earth and the Sun). We noted at that point that the only known exception to this was the case of decaying nuclides whose bonding electrons are those directly involved in the radioactive decay, as specifically in the case of EC and especially as in the case of the EC-decay of ^{7}Be. Now, as we finish this thesis, it appears that we must return again to the same conclusion as that drawn by the Curies, Kammerlingh-Onnes and others, almost one century ago. For it indeed appears, and we have in this paper endeavored to show beyond any doubt that this is so, that truly it may be said that radioactive half-lives, with the exceptions noted above, *do not depend on external environment.*

We believe that our results, accurate to 0.04 - 0.1%, have definitely eliminated the possibility that beta-decay half-life can be altered by amounts greater than this as a result of changes in temperature [15, 16] (see also [115]), or host material [17], or by changes in the Earth-Sun distance [116]. We therefore state with new-found assurance that radioactive half-lives are indeed, as stated so long ago by Curie and Onnes, independent of temperature and whether the decay takes place in a metallic or non-metallic environment.

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