

Systematics of alpha decay half-life: new evaluations for alpha-emitter nuclides

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Abstract - A semiempirical model based on the quantum mechanical tunnelling mechanism of alpha emission from nuclei has been used to systematize the alpha decay half-lives of a set of 336 nuclides, comprising all the alpha-emitter nuclides whose $T_{1/2_\alpha}$ -data for ground-state to ground-state transitions of mutual angular momentum $\ell = 0$ are known. With a minimum of data rejection (only $\sim 5\%$ of cases), the procedure has been successful in reproducing quite satisfactorily (within a factor ~ 2) most of the cases ($\sim 80\%$) investigated. The few significant discrepancies found between measured and calculated results are analysed and discussed. Also reported is the prediction from the model for possible new alpha-emitter nuclides, namely ^{180}W , ^{184}Os , and ^{228}Ra for which cases the calculated partial alpha decay half-lives fall within the range of half-lives measurable by the current techniques.

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Since the pioneer investigation by Geiger and Nuttall in 1911 concerning the relationship of the partial alpha decay half-life with alpha particle energy for a number of radioelements [1], and the first description given independently by Gamow [2] and Condon and Gurney [3] of the alpha decay process as a quantum tunnelling mechanism through a Coulomb potential barrier, a large number of phenomenological (semiempirical in nature) formulae of practical use has been proposed in predicting for alpha decay cases not yet measured [4–8] and/or searching for stability of superheavy elements [8] as well as very neutron deficient nuclei against alpha emission [6]. In addition, detailed theoretical approaches giving microscopic descriptions of the mechanism of alpha decay [6, 9, 10] are still of continuous interest in view of a number of recent discoveries related to alpha decay process [11–16].

Recently, a research group detected for the first time alpha particles from the radioactive decay of natural bismuth (100% of ^{209}Bi isotope) by means of the scintillating bolometer technique with massive BGO ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) crystals [17]. The measured half-life was found as $(1.9 \pm 0.2) \times 10^{19}$ y, corresponding to an alpha activity equivalent to ~ 12 disintegrations/h·kg, which represents the lowest natural alpha activity ever observed. The precise and accurate half-life value obtained in such spectacular experiment motivated us to develop a one-parameter model for the alpha decay process, based on the quantum mechanical tunnelling mechanism of penetration through a potential barrier, where the centrifugal and overlapping effects are taken into account [18]. This treatment enabled us to calculate and systematize the alpha decay half-lives of ground-state to ground-state transitions of mutual angular momentum $\ell = 5$ for all the possible alpha-emitter bismuth isotopes which can be produced by nuclear reactions. It was verified that the measured partial alpha decay half-life-values (covering 25 orders of magnitude) were reproduced by the proposed method quite satisfactorily (in some cases within 15%). In particular, the alpha decay half-life for the naturally occurring ^{209}Bi isotope was evaluated as $(1.0 \pm 0.3) \times 10^{19}$ y [18], in substantial agreement with the experimental result $((1.9 \pm 0.2) \times 10^{19}$ y [17]).

More recently, this semiempirical model of alpha emission from nuclei has been used

also to evaluate the half-life of the Pt isotopes [19]. For the important naturally occurring ^{190}Pt isotope, the radiogenic parent in the $^{190}\text{Pt} \rightarrow ^{186}\text{Os}$ dating system, the model yielded a half-life value of $(3.7 \pm 0.3) \times 10^{11}$ y. This is comparable to $(3.2 \pm 0.1) \times 10^{11}$ y which was obtained in the last direct counting experiment to measure the alpha activity of ^{190}Pt [20].

The proposed model has shown to be successfully applicable also to other isotopic sequences of alpha-emitter nuclides [18, 19]. This has stimulated us to extend its application to all nuclides whose alpha decay half-lives have already been determined experimentally, and also to investigate its usefulness as a tool in predicting alpha-decay half-lives not yet measured. In brief, the decay constant, λ , is calculated as

$$\lambda = \lambda_0 S_\alpha P_{\text{se}}, \quad S_\alpha = e^{-G_{\text{ov}}}, \quad P_{\text{se}} = e^{-G_{\text{se}}}, \quad (1)$$

where λ_0 is the frequency factor which represents the number of assaults on the barrier per unit of time, S_α is the alpha particle preformation probability at the nuclear surface (also known as the spectroscopic factor), G_{ov} is Gamow's factor calculated in the overlapping barrier region where the alpha particle drives away from the parent nucleus until the configuration at contact is reached (the quantity $S_\alpha = e^{-G_{\text{ov}}}$ can, therefore, be thought as the "arrival" of the alpha particle at the nuclear surface), P_{se} is the penetrability factor through the external barrier region (*i.e.*, the Coulomb plus centrifugal potential barriers), and G_{se} is Gamow's factor calculated in this external, separation region which extends from the contact configuration up to the separation point where the total potential energy equals to the Q_α -value for decay. The quantity λ_0 is evaluated usually as $\lambda_0 = \frac{v}{2a}$, where v is the relative velocity of the fragments, and $a = R_p - R_\alpha$ is the difference between the radius of the parent nucleus and the alpha particle radius.

In cases for which the alpha transition goes from the ground-state of the parent nucleus to the ground-state of the daughter nucleus, and the transition has mutual angular momentum $\ell = 0$, and expressing lengths in fm, masses in u, and energies in MeV, the expressions for G_{ov} and G_{se} reduce to (the general expressions when $\ell \neq 0$ are given in [18]):

$$G_{\text{ov}} = g G'_{\text{ov}}, \quad G'_{\text{ov}} = 0.4374702(c - a) (\mu_0 Q_\alpha)^{1/2} \left(\frac{1}{z} - 1 \right)^{1/2}, \quad (2)$$

$$G_{\text{se}} = 1.25988372 Z_{\text{d}} \left(\frac{\mu_0}{Q_{\alpha}} \right)^{1/2} \{ \arccos(z^{1/2}) - [z(1-z)]^{1/2} \}, \quad (3)$$

in which

$$z = \frac{cQ_{\alpha}}{2Z_{\text{d}}e^2}, \quad e^2 = 1.4399652 \text{ MeV} \cdot \text{fm}. \quad (4)$$

Here, $c = R_{\text{d}} + R_{\alpha}$ is the separation of fragments in the contact configuration (R_{d} represents the radius of the daughter nucleus), Z_{d} is the atomic number of the daughter nucleus, and μ_0 represents the reduced mass of the disintegration system. The quantity $c - a$ represents, therefore, the extension of the overlapping region. Both the values of μ_0 and Q_{α} have been calculated from the most recent atomic mass-excess (ΔM) evaluation [21], and they include the effect of the screening to the nucleus caused by the surrounding electrons. Accordingly, the expressions for μ_0 and Q_{α} read:

$$\frac{1}{\mu_0} = \frac{1}{m_{\text{d}}} + \frac{1}{m_{\alpha}}, \quad (5)$$

$$m_{\text{d}} = A_{\text{d}} + \frac{\Delta M_{\text{d}}}{F} - \left(Z_{\text{d}} m_{\text{e}} - \frac{10^{-6} k Z_{\text{d}}^{\beta}}{F} \right), \quad (6)$$

$$Q_{\alpha} = \Delta M_{\text{p}} - (\Delta M_{\text{d}} + \Delta M_{\alpha}) + 10^{-6} k \left(Z_{\text{p}}^{\beta} - Z_{\text{d}}^{\beta} \right), \quad (7)$$

where $m_{\alpha} = 4.001506179144 \text{ u}$ is the alpha-particle mass, $m_{\text{e}} = 0.548579911 \times 10^{-3} \text{ u}$ is the electron rest mass, $F = 931.494009 \text{ MeV/u}$ is the mass-energy conversion factor, and $\Delta M_{\alpha} = 2.42498783 \text{ MeV}$. The quantity kZ^{β} represents the total binding energy of the Z electrons in the atom, where the values $k = 8.7 \text{ eV}$ and $\beta = 2.517$ for nuclei of $Z \geq 60$, and $k = 13.6 \text{ eV}$ and $\beta = 2.408$ for $Z < 60$ have been derived from data reported by Huang *et al.* [22].

As explained in our previous work [18], the value for the equivalent sharp radius of the alpha particle, also adopted in the present systematics, is $R_{\alpha} = (1.62 \pm 0.01) \text{ fm}$ as it comes from the charge density distribution resulting from data on elastic electron scattering from ${}^4\text{He}$ as measured by Sick *et al.* [23]. Finally, the nuclear radii for the parent, R_{p} , and daughter, R_{d} , nuclei have been evaluated following the droplet model of

atomic nuclei [24], from which the following radius expressions have been used throughout:

$$R_i = \frac{Z_i}{A_i} R_{pi} + \left(1 - \frac{Z_i}{A_i}\right) R_{ni}, \quad i = \mathbf{p}, \mathbf{d}, \quad (8)$$

where the R 's are given by

$$R_{ji} = r_{ji} \left[1 + \frac{5}{2} \left(\frac{w}{r_{ji}}\right)^2\right], \quad j = p, n; \quad i = \mathbf{p}, \mathbf{d}, \quad (9)$$

in which $w = 1$ fm is the difuseness of the nuclear surface, and the r 's represent the equivalent sharp radius of the proton ($j = p$) or neutron ($j = n$) density distribution. These latter quantities, in turn, are calculated following the Finite-Range Droplet Model description of nuclei by Möller *et al.* [24], thus giving

$$r_{pi} = r_0 (1 + \bar{\epsilon}_i) \left[1 - \frac{2}{3} \left(1 - \frac{Z_i}{A_i}\right) \left(1 - \frac{2Z_i}{A_i} - \bar{\delta}_i\right)\right] A_i^{1/3}, \quad (10)$$

$$r_{ni} = r_0 (1 + \bar{\epsilon}_i) \left[1 + \frac{2}{3} \frac{Z_i}{A_i} \left(1 - \frac{2Z_i}{A_i} - \bar{\delta}_i\right)\right] A_i^{1/3}, \quad (11)$$

where

$$\bar{\epsilon}_i = \frac{1}{4 e^{0.831 A_i^{1/3}}} - \frac{0.191}{A_i^{1/3}} + \frac{0.0031 Z_i^2}{A_i^{4/3}}, \quad (12)$$

$$\bar{\delta}_i = \left(1 - \frac{2Z_i}{A_i} + 0.004781 \frac{Z_i}{A_i^{2/3}}\right) \left/ \left(1 + \frac{2.52114}{A_i^{1/3}}\right)\right., \quad (13)$$

$r_0 = 1.16$ fm, and, as before, $i = \mathbf{p}$ (parent) or \mathbf{d} (daughter).

Therefore, the standard form for the partial alpha decay half-life formula (expressed in s) is written as

$$\tau = \log_{10} T_{1/2\alpha} = -22.0 + \log_{10} a + \frac{1}{2} \log_{10} \left(\frac{\mu_0}{Q_\alpha}\right) + (g G'_{ov} + G_{se}) \log_{10} e, \quad (14)$$

where g is the adjustable parameter of the model, the value of which being thus determined from experimental data of partial alpha decay half-life of the alpha emitter nuclides included in the systematics.

The above method has been used to systematize the partial alpha decay half-lives of all alpha emitter nuclides. These amount to 336 alpha decay cases which have experimental indication of total half-life and alpha-decay branching ratio for ground-state to ground-state transitions of mutual angular momentum $\ell = 0$ and, therefore, their partial alpha decay half-lives ($T_{1/2}^e$) are known [21, 25]. As a first step, these experimental $T_{1/2}^e$ -values were used as input into equation (14) to get an insight on the individual g -values that would be required to reproduce the corresponding half-life values within the context of the present model. Such an analysis showed that a unique g -value would be sufficient to fit all data, *i.e.* the same g -value is valid for even-even, even-odd, odd-even, and odd-odd parent nuclei. Six of the 336 half-life measurements have been discarded (these were for ^{110}Te , ^{151}Gd , ^{174}Hf , ^{163}Ta , ^{203}Ra , and ^{250}Cm parent nuclei) because their associated g_{exp} -values were found negative, and, according to the details of the model [18], the values of parameter g are physically restricted to the interval $0 < g \leq 2/3$. The remaining 330 nuclides were then used to obtain the best g -value by minimizing the quantity

$$\sigma = \left\{ \frac{1}{N-2} \sum_{i=1}^N \left[\log_{10} \left(\frac{T_{1/2_i}^e}{T_{1/2_i}^c} \right) \right]^2 \right\}^{1/2}, \quad (15)$$

where $T_{1/2}^c$ is the calculated partial alpha decay half-life (equation (14)), and N is the number of alpha disintegration cases considered ($N = 330$). This process is illustrated in Fig. 1(a). The g -value so obtained is then used back into the routine calculation of the model to calculate the $T_{1/2_\alpha}$ -values for the N alpha emitter nuclides. A second data rejection has eliminated ten further $T_{1/2}^e$ -values, namely, the cases for ^{113}I , ^{210}Po , ^{200}At , ^{206}At , ^{208}At , ^{211}At , ^{218}At , ^{231}Pa , ^{266}Sg , and ^{272}Rg , for which cases $|\Delta\tau(= \log_{10} T_{1/2}^c - \log_{10} T_{1/2}^e)| > 2\sigma$. Finally, the process of minimization (equation (15)) was repeated for the set of 320 remaining alpha decay cases included in the present systematics ($\sim 95\%$ of the original input data). By using this new data set as input to our semiempirical, one-parameter model, we found for the adjustable parameter the value $g = 0.122$, corresponding to a minimum standard deviation (equation (15)) of $\sigma_{\text{min}} = 0.317$. It is worth mentioning that the single g -value found in this systematic analysis is comparable (within $\sim 7\%$) to the one obtained very recently for the platinum isotopes alone ($g = 0.131$ [19]).

Figures 1 and 2 summarize the results of application of the present model to systematize the partial $T_{1/2\alpha}$ -values already measured for a number of nuclides. Both figures refer to the final set of 320 nuclides. Figure 1(a) illustrates the process of minimization leading to $g = 0.122$ and $\sigma_{\min} = 0.317$. Figure 1(b) is the frequency distribution of the individual experimental g -values, g_{exp} , which come from equation (14)), and its corresponding gaussian fit giving $\bar{g}_{\text{exp}} = 0.121$. The vertical dashed line clearly shows that the average value \bar{g}_{exp} and the best g -value from part (a) practically coincide, as one should expect from an analysis of a very large number of alpha decay cases.

In Fig. 2 the quantity $\Delta\tau = \tau_c - \tau_e$ is plotted *versus* mass number of the parent nucleus. The figure shows that, despite the large range covered by both the half-life (~ 30 orders of magnitude) and atomic number ($Z = 52-116$) quantities, agreement between calculated and measured values can be considered rather satisfactory (in 26 cases, the difference between each other is indeed very small ($< 10\%$)). The good reproducibility of the alpha decay half-lives achieved by the present model is also illustrated by the frequency distribution of $\Delta\tau$ (at the right), which is well centered very close to zero ($\overline{\Delta\tau} = 0.002$) and shows that the experimental half-lives of about 80% of the nuclides are reproduced within a factor ~ 2 .

In Table 1 we compare the statistics of the present analysis with those we have obtained from other simple, proposed formulae of partial alpha decay half-life [26, 6]. To a correct comparison we used the same input data (Q_α - and $T_{1/2\alpha}^e$ -values) as was done in the present systematics. The values of σ and $\overline{\Delta\tau}$ resulting from the present formalism (last line in Table 1) are better than, or comparable to, the ones obtained from other formulae. A case that is similar to ours (although the input data were not exactly the same as for the present analysis) is the one of Ref. [10], in which case a two-parameter model was used to systematize the half-life measurements of 324 nuclides, leading to a $\overline{\Delta\tau}$ -value slightly better than ours. Concerning the σ -value, however, our systematics is clearly on a better position. Considering that our formalism uses only one parameter to fit all the data, one can say, therefore, that the systematics here described is as good as, or even better than, the ones listed in Table 1.

Finally, as an application of the present systematics, we analyze the half-life for some selected nuclides, the calculated $T_{1/2_\alpha}$ -values of which are listed in Table 2 (the corresponding experimental results are included when available). In Group I we list three naturally occurring nuclides, ^{180}W , ^{184}Os , and ^{228}Ra whose partial alpha decay half-lives have not yet been measured. The prediction of the present model is shown in the fifth column, namely 1×10^{18} y, 2.6×10^{13} y, and 4.6×10^9 y for the tungsten-180, osmium-184, and radium-228, respectively. These figures, which are within the range of half-lives measurable with current techniques, suggest that an attempt should be made in trying to determine these half-life-values experimentally. This is particularly true in the case of radium-228, for which case the calculated branching ratio of $1.2 \times 10^{-7}\%$ makes it the best candidate (among the three) to having its natural alpha radioactivity experimentally detected. In Group II we list the six nuclides (^{110}Te , ^{151}Gd , ^{174}Hf , ^{163}Ta , ^{203}Ra , and ^{250}Cm) whose partial alpha decay half-life measurements have been discarded from the present systematics because their associated g_{exp} -values have been found negative, therefore, physically not acceptable; their $T_{1/2_\alpha}$ -values calculated with the g -value of the present systematics, $g = 0.122$, are, therefore, consistently greater than the corresponding measured values. We remind, from equations (2)–(14), that a negative g -value would be suggestive of a potential barrier (in the overlapping region) favoring the escaping of an alpha-particle from its parent nucleus, which is intriguing. The negative g -values in the six cases discarded, although small in magnitude, are, in our opinion, indicative that new measurements for those partial alpha decay half-lives should be performed. In Group III we list the ten nuclides (^{113}I , ^{210}Po , ^{200}At , ^{206}At , ^{208}At , ^{211}At , ^{218}At , ^{231}Pa , ^{266}Sg , and ^{272}Rg) of measured $T_{1/2_\alpha}$ -values discarded from the present systematics by using the rejection criterion $|\Delta\tau(= \log_{10} T_{1/2}^c - \log_{10} T_{1/2}^e)| > 2\sigma$. In all these cases the measured $T_{1/2_\alpha}$ -values are significantly underestimated (up to two orders of magnitude) by the present model. For the nuclides polonium-210 and astatine-211, a possible explanation for this would be the extra stability they are expected to have, namely an effect of the 126 neutron shell closure on their alpha decay half-lives. In the cases of the alpha emitters astatine-200, astatine-218, seaborgium-266, and roentgenium-272 the authors speculate that their resulting large values of $|\Delta\tau|$ (last column in Table 2) could be associated with

difficulties in performing accurate measurements of these short, or extremely short half-lives, a factor that may play an even more important role in the case of the artificially produced alpha emitters seaborgium-266 and roentgenium-272 (this latter with a reported experimental $T_{1/2\alpha}$ -value of the order of 1 ms). Concerning the remaining four nuclides the authors cannot, at this point, identify a possible explanation for a discrepancy of 1–2 orders of magnitude.

In summary, in the present work the authors carry out a systematization of the partial alpha decay half-lives for all the 336 cases of alpha-emitter nuclides whose $T_{1/2\alpha}$ -values for ground-state to ground-state transitions of mutual angular momentum $\ell = 0$ are known. The systematics uses a one-parameter model described earlier [18, 19], in which the alpha decay process is based on the quantum mechanical tunnelling mechanism of penetration through a potential barrier, where the centrifugal and overlapping effects are taken into account. This treatment enabled us to calculate and systematize the partial alpha decay half-lives of 320 cases after a minimum of data rejection (only 5% of cases). The systematics provides, in our opinion, a quite satisfactory description for the set of 320 remaining alpha decay cases, whose experimental $T_{1/2\alpha}$ -values are reproduced as follows: about 95% of cases within a factor ~ 3 , 80% within a factor ~ 2 , and 26 cases with an accuracy less than 10%. The model has also been used to predict the partial alpha decay half-life for a few nuclides for which this quantity has not yet been measured. For these cases, the authors propose that attempts should be made aiming at the experimental determination of their natural alpha radioactivity. In addition, for at least some of the nuclides whose $T_{1/2\alpha}$ -values cannot be reproduced with the present systematics, the authors suggest that researchers perform further experimental work.

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Table 1 - Statistics obtained in different alpha-decay half-life systematics^a.

Reference for alpha decay half-life formula or formalism	Number of		
	adjustable parameters n	Standard deviation ^b σ	Mean value ^c $\overline{\Delta\tau}$
Alex Brown [26]	3	0.461	0.020
Royer [6]	3	0.298	-0.004
Tavares <i>et al.</i> [10]	2	0.380	0.001
Present work	1	0.317	0.002

^a Except for the case in Ref. [10], the same 320 Q_{α} - and $T_{1/2\alpha}$ -values have been used as input data in the different systematics.

^b The quantity analysed is $\tau = \log_{10} T_{1/2}$.

^c $\overline{\Delta\tau} = \frac{1}{N} \sum_{i=1}^N (\tau_c - \tau_e)$

Table 2 - Partial alpha decay half-life evaluation and comparison with measured values (when available) of some alpha-emitter nuclides.

Group of nuclides	Parent Nucleus		Partial alpha decay half-life (s)		
	Z	A	$T_{1/2}^e$	$T_{1/2}^c$	$\log_{10}(T_{1/2}^c/T_{1/2}^e)$
I ^a	74	180	—	3.16×10^{25}	—
	76	184	—	8.20×10^{20}	—
	88	228	—	1.45×10^{17d}	—
II ^b	52	110	6.20×10^5	2.44×10^6	0.59
	64	151	1.07×10^{15}	7.23×10^{15}	0.83
	72	174	6.31×10^{22}	1.20×10^{24}	1.28
	73	163	5.30×10^3	5.40×10^4	1.01
	88	203	4.00×10^{-3}	3.22×10^{-2}	1.47
	96	250	2.78×10^{12}	1.79×10^{13}	0.81
III ^c	53	113	1.99×10^9	2.00×10^7	-2.00
	84	210	1.19×10^7	1.48×10^6	-0.91
	85	200	3.09×10^2	2.24×10^1	-1.14
			2.06×10^5	2.08×10^4	-1.00
			1.10×10^6	8.84×10^4	-1.10
	211		6.21×10^4	6.28×10^3	-1.00
			4.17×10^1	1.01×10^0	-1.62
	91	231	9.40×10^{12}	9.75×10^{10}	-1.98
	106	266	6.18×10^1	5.74×10^0	-1.03
111	272	2.00×10^{-3}	4.35×10^{-5}	-1.56	

^a Naturally occurring, not yet measured.

^b Cases for which the g_{exp} -values resulted negative.

^c Cases rejected by the criterion $|\log_{10}(T_{1/2}^c/T_{1/2}^e)| > 2\sigma$.

^d This corresponds to a branching ratio of $1.2 \times 10^{-7}\%$.

Figure Captions

Fig. 1 (a) Minimization of the standard deviation σ (equation (15)) to determine the best value of the adjustable parameter of the model, g (equation (2)). (b) Frequency distribution (grey line) of the experimental g -values, g_{exp} (calculated from (14)), and its corresponding gaussian fit (black curve); the vertical dashed line clearly indicates the excellent agreement between the average value \bar{g}_{exp} and the best g -value from part (a).

Fig. 2 The quantity $\tau_c - \tau_e = \log_{10} T_{1/2}^c - \log_{10} T_{1/2}^e$ versus mass number A for the 320 alpha-emitter nuclides included in the present systematics. The frequency distribution at the right emphasizes the good reproducibility of the alpha decay half-lives achieved by the present model (the experimental half-lives of about 95% of the cases are reproduced within a factor ~ 3).

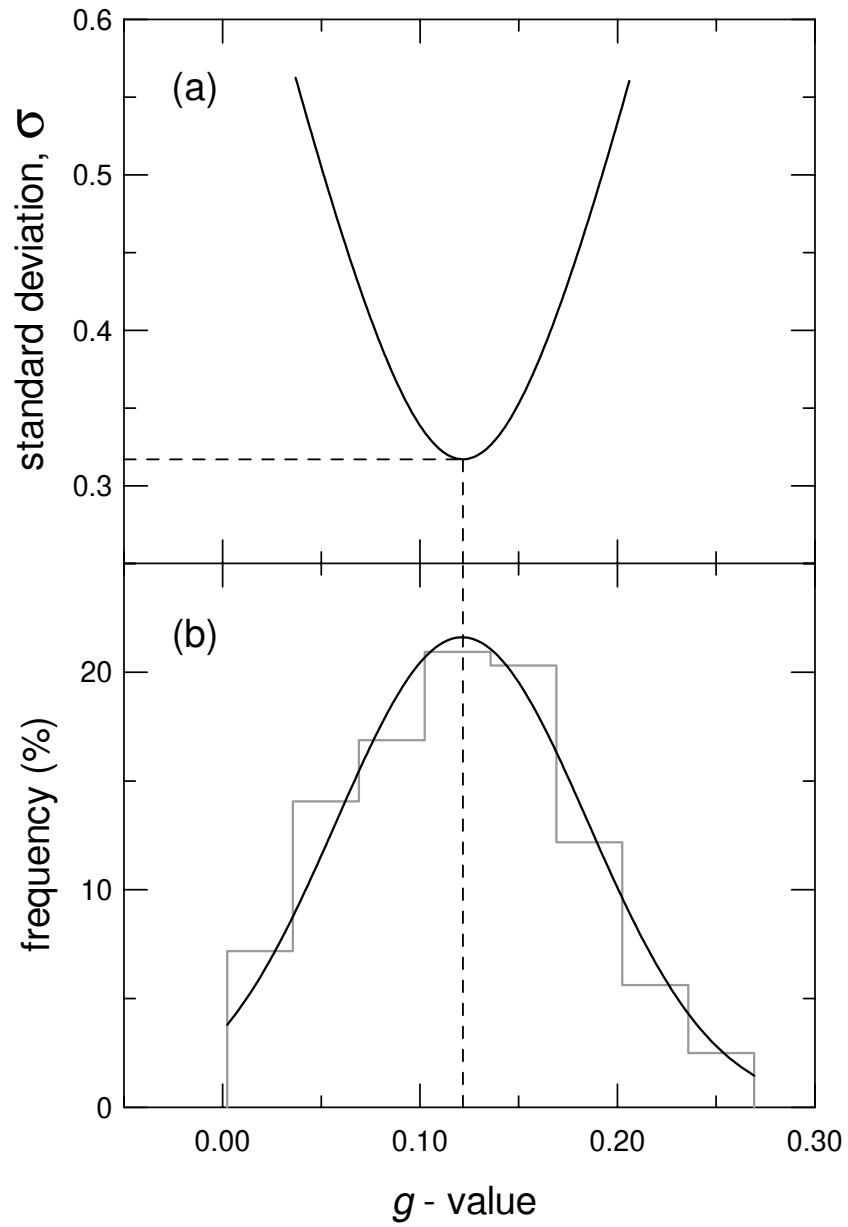


Figure 1

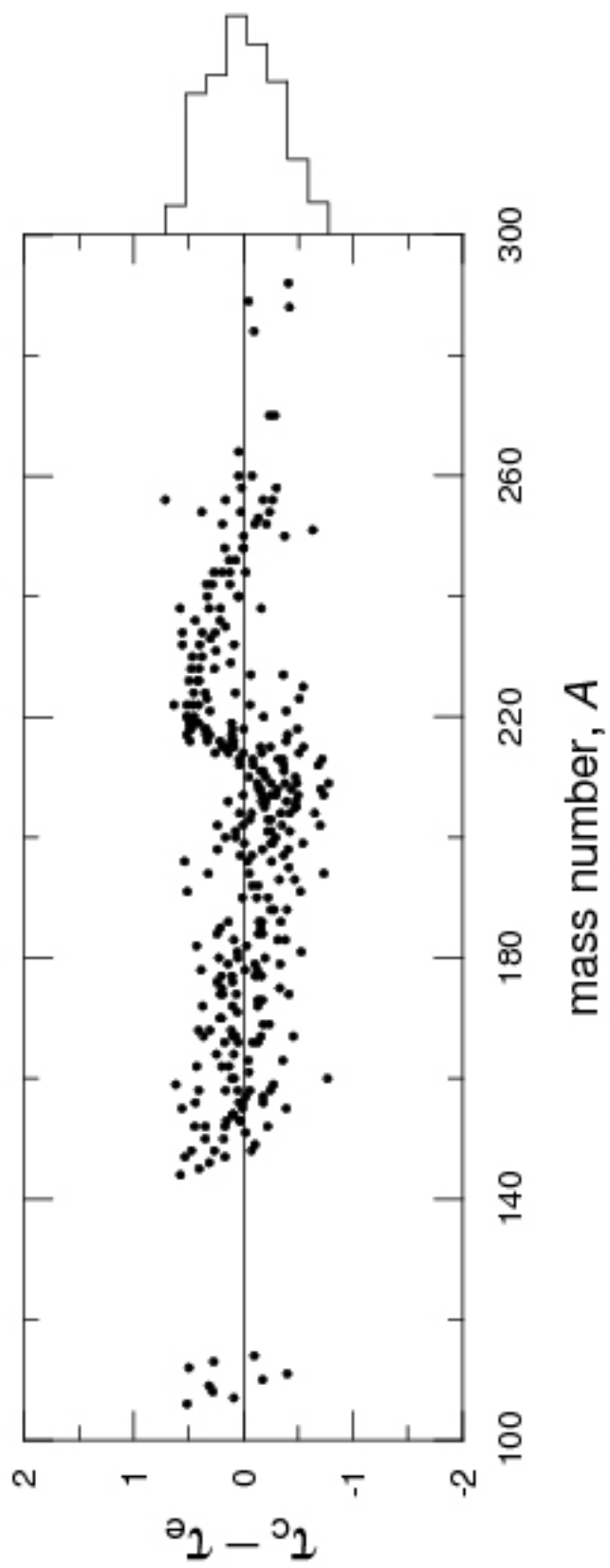


Figure 2