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Limitations on the precision of atomic meanlife measurements

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Abstract

Uncertainty estimates in atomic meanlife measurements are usually obtained by statistical inference based on the hypothesis that the exponential decay law is exact and the distribution of random errors about it is Gaussian. At some level of precision these parameter evaluation methods must be accompanied by additional hypothesis testing. Possible limitations on these accuracies are examined in the light of recent measurements and calculations, and are quantitatively studied through the simulation and fitting of several alternative models. © 1997 Published by Elsevier Science B.V.

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1. Introduction

The estimation of the uncertainties associated with the determination of atomic meanlives from measured decay curves is usually based on the assumption that the fundamental process is exactly governed by the exponential decay law and that the distribution of the random measurement uncertainties is exactly Gaussian. As measurement accuracies continue to increase there must exist a level of precision above which these error estimation procedures must be reexamined, and the parameter evaluation by statistical inference must be accompanied by robust distribution-independent hypothesis testing. We examine these questions in the light of recent measurements and calculations, consider possible uncertainties through the simulation and fitting of several alternative models, and investigate methods for testing the accuracy of quoted uncertainties.

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2. The exponential model

Although the exponential decay law is not a rigorous consequence of quantum mechanics, but rather the result of somewhat delicate approximations [1–6], there is a great deal of empirical support for its validity in tests [7–10] over a wide range of time scales at a precision level of a few percent. However, on a time scale commensurate with that of the meanlife itself, there exists neither theoretical nor experimental evidence that precludes distortions superimposed on the exponential behavior at the level of parts per thousand.

2.1. Tests of exponential decay in nuclear and particle physics

Much of the confidence in the exponential law for the mathematical description of unstable particles is derived from studies of nuclear and fundamental particle decay, where the process is essentially indepen-

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dent of the external environment. Experimental tests of the exponentiality of the rate of decay of these particles have been conducted, based on the constancy of the slope of the semilogarithmic decay curve (see Refs. [3,5–10] and the bibliographies therein). Here (unlike atomic processes), theoretical estimates are usually not of sufficient precision to permit testing of the accuracy of the parameter evaluation. Several recent studies are particularly interesting. One experiment [8] found no deviations from the exponential law during either the first 10^{-4} half-life of ^{60}Co (to within 5.6%), or during the first 45 half-lives of ^{56}Mn (to within 1.3%). Another experiment [9] compared the specific decay rates of a freshly prepared sample of ^{40}K and one that is 4.5×10^9 years old, again with no difference (to within 11%). Yet another experiment [10] tested the exponential decay law for times shorter than 10^{-13} seconds using the decay lengths of tau leptons (to within 8.5%). Nevertheless, although the exponential decay law has been tested on the level of a few percent for absolute times from 10^{-13} to 10^{17} seconds and for decay times from 10^{-4} to 45 half-lives, the degree to which the exponential law is valid on a level of 0.1% for decay times commensurate with the meanlife has not been established either theoretically or experimentally.

In contrast to the tests of the exponentiality of the rate of decay of an unstable sample, another type of test of the exponential law involves a direct measurement of the time dependence of the quantity of the sample which either survives or results from the decay. Recent experiments of this type [11–13], in which the exact validity of the exponential law is assumed, have determined the neutron lifetime with a precision quoted at $\pm 0.2\%$. One type of experiment [11] studied the neutrons that decay (by trapping the proton decay products) and the other type [12] studied the neutrons that survive (by ultracold neutron storage). However, because of the experimental complexities and the large number of parameters involved in the analysis of the data, these experiments do not appear to provide a stringent test of the exponential law.

2.2. *Measurements relating to the validity of the exponential decay model in atomic physics*

In atomic physics the exponential decay law is generally tested only indirectly via the comparison of calculated transition dipole matrix elements with those deduced from lifetime measurements. In this case the tenuous connection between the theoretical transition dipole moment and the predicted mean lifetime makes an exact comparison between the theoretical and experimental values of the lifetime questionable. In particular, since the theoretical transition moment is obtained from a calculation of a transition matrix element between precise energy states (which disallows any information with respect to time), it is only the assumption that the exponential decay law is exact (so that the decay per atom is a constant) that allows the computed transition rate to be equated to the reciprocal of a mean lifetime.

More recently however, Seke [3] and Nicolaides and Mercouris [5,6] have independently provided analyses of decaying states that allow the theory to be more directly related to decay curve measurements. These authors derive solutions to the time-dependent Schrödinger equation which directly determine the time evolution of the given decaying states. In particular, Seke [3] derives an analytic result for the amplitude of the 2p state in hydrogen in the presence of the transition to 1s, while Nicolaides and Mercouris [5] provide a numerical solution to the Schrödinger equation for the time dependence of the $1s2p^2\ ^4P$ autoionizing state in the three-electron He^- system (subsequently extended [6] to the $1s^22s2p\ ^2P^o$ shape resonance in Li^+ and the $3d5p\ ^3F^o$ doubly excited autoionizing state in Ca). The results of these authors reveal the presence of significant nonexponential terms in the time dependent survival probabilities of the decaying states. In particular, the work of Ref. [5] demonstrates that the probability of the initial state is well represented by an exponential only over a limited range of time, which, in the case considered, is equivalent to the first five decades (about 12 meanlives). By fitting the logarithm of the decay curve to a lifetime over this range in their calculation, Nicolaides and Mercouris [5] obtain agreement (to within a few percent) with the width of the state obtained in an earlier calculation [14] of the radiative autoionization cross section. These authors also discuss a proper definition of the lifetime in the case of deviations from the exponential decay law. What seems relevant to the point that we emphasize here is that the results of the authors of Refs. [5] and [3] suggest that neither theory nor experimental analysis

of decay curves can be expected to provide *lifetimes* accurate to the order of 0.1%.

2.2.1. Decay curve measurements

Transition moment calculations can be made to high accuracy for one and two electron systems [15–17]. The mean lifetimes deduced from these calculations can be compared to precision decay curve measurements in the H-like and He-like isoelectronic sequences, which exist to accuracies better than 1%. In particular, lifetime measurements have been made in neutral He for the 1s3p ^{1}P (to 0.26%) [18], 1s3s ^{3}S (to 0.56%) [19], and 1s3d ^{3}D (to 0.42%) [19] levels, and the results of these measurements agree with theoretical calculations to within the quoted accuracies. Measurements have also been made for the He-like 1s2s ^{3}S levels in C^{4+} (±0.23%) [20] and Nb^{39+} (±10.35%) [21], but here theoretical calculations are not yet of the same precision.

Another system with a simple structure which allows for a study of the decay process is the positronium atom. In this case two very precise (±0.02%) measurements of the decay curve of the 1s ^{3}S ground state [22–24] have been made, but these differ significantly (by 0.19% and 0.12%) from QED calculations [25,26].

For favorable cases in more complex atoms, present experimental methods permit atomic meanlives to be extracted from decay curve measurements to within accuracies better than parts in 10^3 [27–29]. In cases where the atoms can be modeled as few electron systems, transition dipole moments can be calculated to similar accuracies [31,32], and the results can be compared with measured meanlives. These comparisons have been found to be acceptable where the achievable experimental accuracies are 2–5%, but as the quoted experimental accuracies have been improved to better than ±0.5%, troubling discrepancies have emerged. In particular, small but significant differences have been found to exist between theoretical calculations (e.g. [31,32]) and experimental measurements [27–29] for the lifetimes of the lowest resonance transitions in the lithium and sodium atoms. For the 2p $^2P_{1/2}$ level in neutral Li, the lifetime measurement with the highest quoted accuracy (±0.13%) disagrees with precision (±0.0001%) theoretical calculations (by 0.7%). Since these atoms possess a relatively simple structure for theoretical consideration and have strong emission lines which allow for selective population in experimental studies, they provide a benchmark test, and the origin of these discrepancies has been eagerly sought. A recent measurement [30] removed an earlier discrepancy [27] for the 3p lifetimes in nuetral Na.

2.2.2. Measurements not involving decay curves

Recently, an alternative experimental method has been applied to the Li [33], Na [34] and K [35] atoms, in which the value of the corresponding transition dipole moment is extracted from molecular spectra without recourse to the exponential decay law. The quoted accuracy of these measurements is very high (±0.02% for Li [33], ±0.1% for Na [34], ±0.2% for K [35]), and they agree well with theoretical calculations [31,32,36,37]. In addition, a high precision measurement (±0.26%) of the H-like He^+ 2p transition matrix element has been made [38] by an alternative electric field quenching method that does not involve the exponential decay law, and this also agrees well with theoretical calculations.

These agreements between theoretical and experimental values of transition dipole moments, and the contrasting discrepancies that occur in some cases when these values are connected to meanlives, raises additional questions concerning the accuracies that can be obtained by the analysis of decay curves.

3. Modeling studies

Our purpose here is not to enquire into the exactness of the exponential decay law, but rather to examine the degree to which factors in addition to the estimated systematic and random errors associated with the measurement could contribute to uncertainties in the experimental meanlife. We have therefore considered four models in which a level decays with an intensity $I(t)$ as a function of time t with an exponential meanlife τ that is distorted by a small modification. These include:

(a) a sinusoidal modulation

$$I(t) = I(0)\exp(-t/\tau)[1 + C\sin(2\pi/T)], \qquad (1)$$

which could result from possible excitation coherences;

(b) a logistic curve

$$I(t) = \frac{I(0)\exp(-t/\tau)}{1 - C[1 - \exp(-t/\tau)]}, \tag{2}$$

which is the solution to the population equation when modified by the addition of a term that is quadratic in the population [39];

(c) a "Khalfin" reciprocal power law tail added to the exponential function

$$I(t) = I(0)\left(\exp(-t/\tau) + \frac{C}{1 + (t/\tau)^4}\right), \tag{3}$$

which is predicted by some models [3,40,41];

(d) a low order power law multiplying the exponential function

$$I(t) = I(0)\exp(-t/\tau)(1 + Ct/\tau), \tag{4}$$

which is also predicted by some models [4,42].

The expressions in each of the above models, as well as the pure exponential decay model (for which $C = 0$), were used to generate arrays of decay curve intensities $I(t_i)$ at time coordinates t_i. These were then modified to simulate experimental data $I_g(t_i)$ through the point-by-point introduction of random Gaussian errors,

$$I_g(t_i) = I(t_i) + \sqrt{I(t_i)}G^{-1}(R_i), \tag{5}$$

where $G^{-1}(x)$ is the inverse Gaussian function and $x = R_i$ is a random number generated between 0 and 1. In order to obtain 0.1% statistical accuracies for $C = 0$, the initial intensities were set at $I(0) = 10^5$, and the decay curves were followed over eight meanlives. Simulated decay curves were generated for each of the four models for various values of the distortion parameter C and the sinusoidal period T (including the pure exponential case $C = 0$).

The decay curves were then fitted by weighted least squares methods (with this statistical accuracy, least squares and maximum likelihood fits are equivalent [43]), and the uncertainties in the fitted lifetimes were estimated by Gaussian statistical inference. In order to compare the individual sample populations with the parent population from which they were selected using Eq. (5), a total of 1000 independent statistical samples were generated for each model. In this manner the average value and variance of the fitted lifetimes and the weighted deviations about the fits could be studied over this large group of statistical samples.

Since the use of an inappropriate fitting function produces a non Gaussian distribution in the deviations from that fit, studies of that distribution can indicate the reliability of the uncertainties predicted by statistical inference. One measure of the distribution is the quantity χ^2_{Red}, which is the sum of the squares of the weighted deviations from the fit divided by the number of degrees of freedom (data points minus fitting parameters) [44]. This quantity characterizes the width of the distribution, but does not probe possible non Gaussian correlated groupings of the deviations. A measure of this correlation can be obtained from the linear correlation coefficient [45]

$$r \equiv \frac{\sum_i x_i y_i}{\sqrt{\sum_j x_j^2}\sqrt{\sum_k y_k^2}}, \tag{6}$$

where x_i and y_i denote paired sets of deviations from an exponential fit. For each model studied in this work we have computed the correlation coefficient r between two different types of paired sets. In what follows we label by r(Ind) the correlation coefficient between corresponding t_i coordinates in two independent statistical samples, and by r(Adj) the correlation coefficient between adjacent t_i coordinates in the same statistical sample.

4. Results

The results are presented in Table 1. Calculations were performed for the values of the distortion parameters C listed (the value of the sinusoidal period chosen here is $T = 2\tau$). With these values, the individual semilog plots of the decay curves were visually indistinguishable from a pure exponential. For the undistorted case and the four distortion models, the fits to a pure exponential had a predicted uncertainty of 0.1%, as determined from the error matrix under the assumption of Gaussian statistics. Averaged over the 1000 samples, the variance in the fitted values of τ confirmed the predicted uncertainties, but the average extracted value for τ differed from the model value by more than 1% for all four models. Thus, as shown in Table 1, the lifetimes extracted for all four models are inaccurate by 10 standard deviations, with the un-

Table 1
Weighted least squares fitted values of simulated data sets, with statistical errors corresponding to $I(0) = 10^5$. The value of τ is averaged over 1000 statistical samples, "Uncertainty" denotes the uncertainty in each fit inferred by Gaussian statistics, "Variance" is the variance of the fitted τ over the 1000 samples, χ^2_{Red} is the sum of squared weighted deviations from the fit per degree of freedom; $r(\mathrm{Ind})$ is the correlation between two statistically independent samples, and $r(\mathrm{Adj})$ is the correlation between adjacent points within the same sample

Model	C	Fitted τ	Uncertainty	Variance	χ^2_{Red}	$r(\mathrm{Ind})$	$r(\mathrm{Adj})$
pure exponential	0	1.0002τ	0.102%	0.104%	1.0	0.00	0.00
(a) sinusoid	0.05	0.9873τ	0.104%	0.101%	11.6	0.91	0.85
(b) logistic	0.05	1.0126τ	0.104%	0.104%	1.6	0.22	0.21
(c) Khalfin	0.03	0.9897τ	0.101%	0.100%	1.8	0.11	0.12
(d) power law	0.01	1.0100τ	0.105%	0.104%	1.0	0.00	−0.03

certainties estimated both by statistical inference and by the variances observed in repeated simulated measurements.

These simulations thus provide a valuable test of our ability to detect inaccuracies in meanlives obtained from fitted decay curves through studies of the deviations from the fit, as indicated by χ^2_{Red}, $r(\mathrm{Ind})$ and $r(\mathrm{Adj})$. These tests signaled a strongly non Gaussian distribution in the deviations from the exponential fit in the cases of models (a), (b) and (c). As shown in Table 1, the values for χ^2_{Red}, $r(\mathrm{Ind})$ and $r(\mathrm{Adj})$ were largest for the sinusoidal (a) model, produced by the coherent oscillations about the fit that are characteristic of this type of distortion. The values were smaller but significant for the logistic (b) and Khalfin (c) models, but for the multiplicative power law (d) model, these tests were unable to distinguish the fit from that produced by a pure exponential model.

The failure of the tests in the case of the power law can be understood on the basis of the expansion formula for the exponential. Specifically, while the multiplicative power law (d) differs significantly from an exponential over a sufficiently large time scale, the two laws are essentially equivalent on the time scale typified by meanlife measurements, provided only that the distortion parameter C is sufficiently small that $Ct \ll \tau$. In this case $1 + Ct/\tau \simeq \exp(Ct/\tau)$, and the fitting function remains exponential but with a distorted effective meanlife $\tau_F \simeq \tau/(1 - C)$. It is the fact that the exponential fitting function is effectively valid in this case that causes the deviations from the fit to be normally distributed, and prevents the statistical analysis from revealing the error in the determination of τ. From this it follows that a multiplicative power law modification of the exponential decay law could easily lead to a discrepancy between the measured value of a decay lifetime and the value deduced from a calculated transition dipole moment.

5. Conclusions

As the precision of the extraction of effective meanlives from atomic decay curves has increased to parts in 10^3 and better, discrepancies between the dipole transition moments deduced from these measurements and those obtained from theory and by other experimental methods have arisen. The studies presented here indicate that deviations from the validity of the exponential law that are too small to be directly detected could lead to inaccuracies in the value of the extracted meanlives that are significant at this level of precision. Thus, while increasingly high precision may be obtained in the extraction of effective meanlives from measured decay curves, ascribing the same precision to the deduced dipole transition moment requires additional theoretical and experimental justification.

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