Radioactivity

Seeking non-exponential decay

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ONE of the most famous laws of physics is the exponential-decay law of radioactivity[1]: the decay of any radioactive isotope is characterized solely by its time constant τ or equivalently by its half-life $t_{1/2} = \tau \log_e 2$. During a time t, $N_0 \exp(-t/\tau)$ nuclei will decay, where N_0 is the number of radioactive atoms at the beginning of the interval. This law has now been checked by Norman *et al.*[2] for extremely short periods, down to 0.01 per cent of a half-life, and long periods, up to 45 half-lives. In neither case was any deviation from exponential decay found. Why should so much effort be expended testing a law with such a venerable history? Because it is incompatible with the equally venerable theory of quantum mechanics[3].

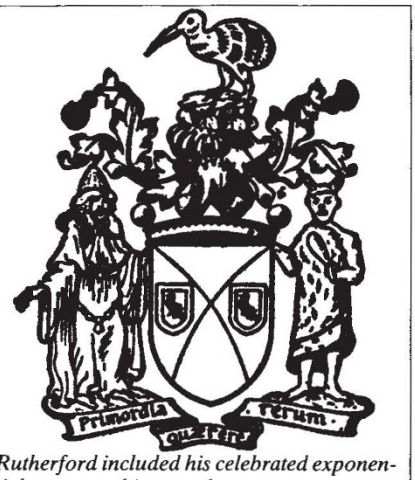
Classically, the exponential law holds because each radioactive atom in an ensemble decays independently and with a probability that is independent of time. As decay proceeds, there are fewer remaining radioactive atoms and the sample's activity falls. Exponential decay is seen in atomic and molecular decay processes as well as in radioactivity.

In quantum mechanics, the decay law of a prepared system should be derived from first principles. The system is described by a state vector Ψ which includes information on how many atoms have already decayed. A 'hamiltonian' operator $\hat{H}$ controls the dynamics of the system, which evolves according to the Schrödinger equation as $\Psi(t) = \exp(-i\hat{H}t)\Psi_i$, where Ψ_i is the state at time $t = 0$.

The decay law is obtained by evaluating the survival probability, that is, the component of the initial state remaining in wavefunction $\Psi(t)$ at time t (denoted $P(t) = |A(t)|^2 = |\langle\Psi_i|\Psi(t)\rangle|^2$). Certain physical restrictions must be put on $\hat{H}$ and Ψ_i (ref. 4). Briefly, there must be a lower limit to the possible energy of the decayed nuclei, and for decay to take place, the spectrum of energies must contain a continuum. This is sufficient to show that the decay rate of any state must ultimately become slower than exponential at very large times. The mean energy of the initial state Ψ_i must be finite, which is sufficient to prove that the initial decay rate of any state must be slower than exponential for small times.

In fact the decay of an isolated quantum state can never be exponential. This is because the products of the decay at an earlier time can recombine to form the initial state later. (This can be seen by subdividing the interval t into two intervals t_1 and t_2. The probability amplitude $A(t)$ must satisfy $A(t_1+t_2) = A(t_1)A(t_2) + \langle\Psi_i|\exp(-i\hat{H}t_2)\,|\Psi_d(t_1)\rangle$, where Ψ_d is the state of the decay products at time t_1. The second term is the 'memory' term that is additional in the quantum-mechanical treatment[5], modifying the classical effect represented solely by the first term.) Thus quantum mechanics implies that the system has a memory, so that it is possible to determine the absolute age of a sample by examining its decay law, which is not possible for pure exponential decay.

In the new experiments, Norman *et al.*[2] searched for deviations from exponential



Rutherford included his celebrated exponential curves on his escutcheon.

decay for short and long times in the radioactivity of ^{60}Co and ^{56}Mn. ^{60}Co, which has a half-life of 5.271 yr, was made from ^{59}Co by neutron irradiation in a pulsed reactor. This defines a precise starting time for the experiment, and Norman *et al.* examined the decay law for the first 10^{-4} half-lives. Similarly, the decay of ^{56}Mn, with a half-life of 2.5785 h, was examined over 45 half-lives, by which time the initial activity had fallen by a factor of about 3×10^{14}. Neither isotope revealed any deviation from exponential decay, and nor has any other test[6]. This seems to be a paradox.

The resolution of this paradox lies in the form that Ψ_i must have if we are to speak of a decaying state. Essentially the decay time τ of Ψ_i must be long in comparison with the characteristic evolution times τ_W associated with the interactions which (nearly) bind the decay products of Ψ_i. Or equivalently, Ψ_i must be constructed from a wavepacket which is much narrower in energy than the range spanned by the interaction responsible for the decay[7]. Under these circumstances the memory time is of the order of τ_W, very much less than the decay time τ, and the memory term then becomes negligible.

Significant deviations from exponential decay occur actually only for very small[8] and very large times. The intrinsic memory time τ_W, and the exponential decay time τ have already been identified. Two other times are important[9]: $\tau_E = \hbar/E_0$ where E_0 is the energy released in the decay and $\hbar$ is Planck's constant, and $\tau_L \sim 3\tau\log_e(E_0\tau/\hbar)$. One always has $\tau_W << \tau$. For decays with very small energy release, however, τ_E increases and τ_L can decrease into regions where observation is possible. It can be shown that exponential decay sets in only after the maximum of τ_E and τ_W and lasts until τ_L, so for decays near threshold ($E_0 \to 0$) there is no exponential decay region[10].

But to see deviations from exponential decay, the decay products must be allowed to recombine. This is not a problem at short times; indeed a straightforward estimate of the time required for the decay products to separate by more than a de Broglie wavelength (the characteristic distance for quantum systems) gives τ_E, but for long times the situation is different. Any fluctuations, including those associated with observation in quantum mechanics, will destroy the recombination effect and therefore produce exponential decay. Furthermore, the decay constant will be τ, the undisturbed value, and independent of the characteristic timescale of the fluctuations if these occur predominantly in the exponential region of the decay [9]. This is likely to prevent the observation of post-exponential decay, particularly in nuclear systems, where τ_L is large. For ^{56}Mn, $\tau_L \approx 3$ weeks, and the decay products must be able to recombine after this time; $\tau_E \approx 10^{-21}$ s so the pre-exponential decay will also not be observed.

Thus it seems unlikely that nuclear decays will show deviations from the exponential-decay law which they made famous. The most promising process for observing non-exponential decay is the near-threshold photodetachment of electrons from negative ions using highly stabilized lasers[11]. In this case, there is a chance of extending τ_E, the length of the pre-exponential region, and of shrinking τ_L, the time of onset of post-exponential decay. The correct combination of circumstances does not seem to arise naturally to produce deviations from exponential decay, which is why it is such an accurately fulfilled law, even out to 45 half-lives. □

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