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Oscillations in radioactive exponential decay

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ABSTRACT

Several older and recent reports provided evidence for the oscillatory character of the exponential decay law in radioactive decay and attempted to explain it with basic physics. We show here that the measured effects observed in some of the cases, namely in the decay of ^{226}Ra, ^{32}Si in equilibrium, and ^{36}Cl, can be explained with the temperature variations.

1. Introduction

Exponential decay law is expressed as the quantum-mechanical probability of survival of a single atom in time t, $e^{-\lambda t}$, where λ is the decay constant, which is equal to the ratio of the average number of atoms that survived the decay to the original number of atoms. The exponential decay law has been satisfied in nuclear systems, whereas evidence exists for deviations from it in atomic systems (see Ref. [1] for a review). Studying exponential decay is fundamentally important since it provides an insight to the decay of quantum state. Exponential decay has wide applications in several fields as it is used for quantification of radionuclides.

Several investigations revealed fluctuations superimposed on average exponential decay curves, which can be seen as clear oscillations in many systems. Alburger et al. measured the decay of ^{32}Si and ^{36}Cl on a gas proportional counter [2]. Both decay curves have showed oscillations superimposed on them with a period of 1 year and a phase shift of approximately 1 month. The maximum occurred in the summer and the minimum in winter. In addition, a ratio ^{32}Si/^{36}Cl was calculated, in which the oscillations are reversed, i.e., they exhibit maximum in winter and minimum in the summer. The relative amplitudes (relative peak-to-valley deviations) of the counting rates were between 1.5×10^{-3} and 3×10^{-3}. The radioactive decay curves for ^{226}Ra and daughters that had been measured in an ionization chamber by Siegert et al., reveal oscillations of similar amplitude, with the maximum in the winter and minimum in the summer [3]. Jenkins et al. correlated the ^{32}Si/^{36}Cl and ^{226}Ra oscillations with the distance between the Earth and the Sun (the shorter the distance in winter, the higher the decay rate measured) [4]. This correlation led them to two possible explanations: solar scalar field modification of the fine structure constant and thus the decay rates or neutrino flux from the Sun interacting with the radioactive nuclei in a novel way. Seasonal oscillations are also seen in the decay of ^{152}Eu measured on a Ge(Li) γ-ray spectrometer by [3].

Godovikov measured the decays of two Mössbauer sources: ^{119m}Sn and ^{125m}Te using a NaI detector [5,6]. It is remarkable that both measurements revealed seasonal oscillations with the same period of 1 y and the same phase shift with maximum in the summer and minimum in the winter. They resemble the seasonal variations for the proportional detector or ionization chamber data [2,3], with the exception of the relative amplitudes be-

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ing much higher (between 0.093 and 0.17). These results were explained by the author with the multiple absorptions and reemissions of γ quanta in the stable Sn and Te nuclei of the source. This was hypothesized as a collective nuclear system in which individual decay events were no longer spontaneous, and thus leading to the oscillations. Opalenko et al. suggested that the oscillations could be caused by instability in the detection system and a narrow discriminator window for the Mössbauer measurements [7].

Litvinov et al. observed oscillations with a period of about 7 s superimposed on the electron capture (EC) decay curves for highly ionized, hydrogen-like ^{140}Pr and ^{142}Pm [8]. The ions were produced by projectile fragmentation and fragment separation at the GSI Darmstadt accelerator, followed by an injection into a cooler-storage ring. The EC decay was detected by change of the ion evolution frequency due to a different mass of the daughter nucleus. Litvinov et al. proposed that the oscillations were caused by energy splitting due to two neutrino mass eigenstates. However, the oscillations were not confirmed for ^{142}Pm, in an experiment by Vetter et al. consisting of a more traditional counting of X rays from the ^{142}Nd daughter [9].

The proposed explanations for the oscillations invoke basic physics and astrophysics. It is, therefore, important to carefully evaluate the data and evidence provided. The purpose of the present Letter is to reanalyze the data by Alburger et al. and Siegert et al., leading to alternative explanations of the oscillations in exponential decay.

2. Ionization chamber data

Siegert et al. reported a nearly 12-y decay data for the ^{226}Ra ($T_{1/2} = 1600$ y) comparison source in conjunction with the half-life measurements of Eu isotopes [3]. Jenkins et al. obtained the original data, extended up to 15 years, from those authors [4]. The background-corrected data exhibit periodic oscillations about a fitted exponential decay with relative amplitude (relative peak-to-valley deviations) of the counting rate $\delta C = 3 \times 10^{-3}$. Siegert et al. suggested that the oscillations could be caused by a discharge on the ionization chamber electrode due to radon daughters in the air, which show seasonal variations. Such oscillations could also be due to background, humidity, or temperature variations.

The ionization-chamber measurement is characterized by a high counting rate from the radium source and a small background. There is no information about the value of background [3], so possible oscillations due to background cannot be evaluated for these data. The humidity could affect the density of the gas in the ionization chamber. However, since the chamber constitutes a closed system, no ambient seasonal humidity variations would affect the gas inside the chamber. Such humidity variations, similarly to the radon daughters, could affect any possible discharge on the electrode.

We have observed gain shifts due to temperature variations in ionization chamber experiments in our laboratory [10]. The effect of temperature on the oscillations can be easily estimated using principles of radiation interaction. The ionization chamber used by Siegert et al. consisted of a 32-cm high cylinder with a 5-cm diameter well for insertion of the source and a 7.2-cm radial length [11]. It was pressurized with argon to $p = 20$ atm (2 MPa). The ^{226}Ra source, in equilibrium with the daughters, was encapsulated in a stainless steel tube. Therefore, γ radiations from the radium daughters were detected in the ionization chamber.

Since the chamber operated at a constant pressure, it follows that $\rho T = \text{const}$, where ρ is the gas density and T an absolute temperature, according to the ideal gas law. Therefore, a small relative temperature change $\delta T = |\Delta T/T|$ changes the relative gas density $\delta\rho = |-\Delta\rho/\rho|$, such that $\delta\rho = \delta T$. The increase of gas density causes greater γ-ray attenuation and, consequently, a higher counting rate. It can be shown that the relative counting rate change $\delta C = |\Delta C/C|$ in the detector due to this effect is given by

$$\delta C = \frac{e^{-\mu\rho x}}{1 - e^{-\mu\rho x}} \mu\rho x \delta T, \tag{1}$$

where μ is an average γ-ray attenuation coefficient and x is the average length of the radiation path in the chamber.

We calculated $\mu = 0.07703\ \text{cm}^2\,\text{g}^{-1}$ in argon using major γ rays from ^{214}Pb and ^{214}Bi [12] and their attenuation coefficients [13]. From the gas law, $\rho/p = \text{const}$, which results in $\rho = 0.03567\ \text{g}\,\text{cm}^{-3}$ for argon at the operating pressure [14]. Since the ion chamber height was greater than the radial length, we set $x = 14.4$ cm, double the radial length, as an average path of the γ rays. With these values, we calculated from Eq. (1) that the observed $\delta C = 3 \times 10^{-3}$ can be caused by $\Delta T = 0.91\,^\circ\text{C} = 1.6\,^\circ\text{F}$.

The above result must be considered as a lower limit. In addition to γ rays, Compton scattered photons from γ interactions in the stainless steel container will reach the ionization chamber. Also, bremsstrahlung X rays are emitted from the β particle interactions in the stainless steel container. It is difficult to calculate this effect, owing to a continuous β-energy spectrum; however, the average X-ray energy is expected to be lower than the average γ-ray energy resulting in higher μ and larger δT in Eq. (1). Nevertheless, the intensity of the X rays is expected to be lower than that of the γ rays.

Siegert et al. did not report seasonal temperature variations ΔT in their laboratory. However, it is likely that, in a temperate climate zone, the average temperature in an institutional building is higher in the summer than in the winter, if all factors such as ambient temperature, ventilation, heating, and air conditioning are taken into account.

3. Proportional detector data

Using a β gas proportional detector, Alburger et al. measured a source containing ^{32}Si ($T_{1/2} = 172$ y, $E_{\beta\,\text{max}} = 225$ keV) [2], which decays to ^{32}P ($E_{\beta\,\text{max}} = 1710$ keV, $T_{1/2} = 14$ d) establishing a secular equilibrium. A ^{36}Cl ($T_{1/2} = 3.0 \times 10^5$ y, $E_{\beta\,\text{max}} = 709$ keV) comparison source was also measured. The 4-y decay data exhibit individual seasonal counting rate oscillations for the two sources, with the maximum in the summer and minimum in the winter. The ratio (^{32}Si + ^{32}P)/^{36}Cl of the normalized counting rates (abbreviated as Si/Cl) was calculated to cancel out any instrumental factors. The ratio exhibits the oscillations in an opposite direction, with the maximum in the winter and minimum in the summer. The extent of the seasonal oscillations is reproduced in Table 1. It is seen that the δC for the Si/Cl ratio, taken between maximum and minimum data points, is about 2.7×10^{-3} (rows 1 and 3), whereas an empirical fit to the data provided δC of 1.5×10^{-3} (row 2).

The ^{32}Si + ^{32}P source (abbreviated as Si) was incorporated in a SiO_2 matrix with an Al absorber on the top, for a total mass thickness of 17 $\text{mg}\,\text{cm}^{-2}$ [2], which considerably limited the contribution from ^{32}Si due to self-absorption of β particles, and emphasized the contribution from ^{32}P, to the observed β counting

Table 1
Observed relative amplitudes δC of the Si and Cl counting rate oscillations.

Parameter	δC	Description	Ref.
Si/Cl	2.7×10^{-3}	max to min data	[2, Fig. 4]
Si/Cl	1.5×10^{-3}	empirical fit	[2, Fig. 4]
Si/Cl	2.8×10^{-3}	max to min data, 5 pt average	[4, Fig. 2]
Cl	4.3×10^{-3}	max to min data	[2, Fig. 3]
Si	2.8×10^{-3}	max to min data	[2, Fig. 3]
Si/Cl	1.5×10^{-3}	δC(Cl)–δC(Si)	[2, Fig. 3]