

Nuclear decay anomalies as a signature of axion dark matterXin Zhang^{1,2}, Nick Houston^{3,*} and Tianjun Li^{4,5}¹*National Astronomical Observatories, Chinese Academy of Sciences,
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A number of nuclear decay anomalies have been reported in the literature, which purport to show periodic variations in the decay rates of certain radioisotopes. If these reports reflect reality, they would necessitate a seismic shift in our understanding of fundamental physics. We provide the first mechanism to explain these findings, via the misalignment mechanism of QCD axion dark matter, wherein oscillations of the effective θ angle induce periodic variation in nuclear binding energies and hence decay rates. As we expect this effect to be most pronounced in low- Q systems, we analyze 12 years of tritium decay data ($Q \simeq 18.6$ keV) taken at the European Commission's Joint Research Centre. Finding no statistically significant excess, we exclude axion decay constants below $9.4 \times 10^{12} - 1.8 \times 10^{10}$ GeV (95% confidence level) for masses in the $1.7 \times 10^{-23} - 8.7 \times 10^{-21}$ eV range.

DOI: [10.1103/PhysRevD.108.L071101](https://doi.org/10.1103/PhysRevD.108.L071101)**I. INTRODUCTION**

As a cornerstone of modern physics, it is widely accepted that radioactive decay is in general a truly random process, occurring independently of time, space and external influence. This simple fact carries with it a vast array of consequences, across fields as diverse as modern-day nuclear medicine to the big bang nucleosynthesis which occurred in the first few minutes of our cosmic history.

Nonetheless, in recent decades a number of purported nuclear decay anomalies have been reported in the literature, which represent apparent violations of this rule [1,2]. Generally these anomalies take the form of periodic variations in the observed decay rate, although notably there are some indications of temporary variations in nuclear decay correlated with solar flares and other astrophysical phenomena [3–6].

One economical explanation of these conclusions, supported by a body of evidence, is that they are largely the

result of random noise, unaccounted-for systematic effects and incomplete uncertainty analysis [7–15]. For example, the overall preference of some datasets for an annually modulated signal may be simply attributed to the influence of seasonally varying environmental conditions, rather than any exotic deviations from known physics [14]. It should also be noted that although radioactive decay is of course a relatively well-explored and understood topic, accurately quantifying the various associated uncertainties is nonetheless nontrivial [15].

This perspective may be supported by the fact that such anomalies are furthermore somewhat difficult to explain in the context of ordinary particle physics. Given the annual periodicity of some claimed signals, solar neutrinos are often suggested to be in some way responsible, but there is no known mechanism within the Standard Model which allows this [1]. Some somewhat speculative hypotheses have been proposed, as summarized in [9,16], but at present there appears to be no concrete framework within which these anomalies can be understood consistently with other observations.

Nonetheless, the absence so far of a compelling explanatory framework does not mean that none exist to be found. Furthermore, regardless of the provenance of these unusual observations, and in particular the key question of if they

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indeed represent a signature of new physics, if they can be connected to some favoured model of physics beyond the Standard Model, they can provide novel constraints and experimental strategies.

Bearing these motivations in mind we, in the following, provide the first mechanism to explain this phenomenon, via a class of particles known as axions. Such an approach carries with it the benefit of also addressing certain other problems in fundamental physics, namely the nature of dark matter (DM) and the strong CP problem in the Standard Model.

Arising originally via a minimal extension of the Standard Model; the Peccei-Quinn solution of the strong CP problem [17–19], axions and axionlike particles occupy a rare focal point in theoretical physics, in that they are simultaneously a generic prediction of the exotic physics of string and M -theory compactifications [20–22]. Despite the profound differences between these two contexts the resulting axion properties are also largely universal, providing an easily characterizable theoretical target. As light, long-lived pseudoscalar particles they can also influence many aspects of cosmology and astrophysics, creating a wide variety of observational signatures [23]. In particular, they are a natural candidate for the mysterious DM comprising much of the mass of our visible Universe [24,25], and as such are a topic of intense ongoing investigation [26].

The aforementioned strong CP problem is the question of why the effective QCD θ parameter, which enters via the Lagrangian term

$$\mathcal{L}_\theta = \frac{g^2\theta}{32\pi^2} G_{\mu\nu} \tilde{G}^{\mu\nu}, \quad (1)$$

is smaller than 10^{-10} in absolute value and not $\mathcal{O}(1)$, as would be expected on the grounds of naturalness. Here g is the gauge coupling and $G_{\mu\nu}$ the gluon field strength tensor. Axions which enact the Peccei-Quinn solution of the strong CP problem do so by promoting θ to a dynamical variable, which can then relax to zero as required.

In the event that these axions also comprise the DM in our Universe, the misalignment mechanism ensures that the time-dependent θ angle today is

$$\theta \simeq \sqrt{\frac{2\rho_{\text{DM}}}{m_a^2 f_a^2}} \cos(\omega t + \vec{p} \cdot \vec{x} + \phi), \quad (2)$$

where ρ_{DM} is the DM density, $\omega = m_a(1 + \frac{1}{2}v^2 + \mathcal{O}(v^4))$, p , v , f_a , and m_a are the axion-field momentum, velocity, decay constant and mass, respectively, and ϕ is an arbitrary phase.

Since various aspects of nuclear physics are θ -dependent this has the potential to lead to a variety of observational signatures, from which constraints on the axion-parameter space can then be placed. For example, Refs. [27–30]

search for an oscillating neutron electric dipole moment (nEDM), and in Ref. [31] it was also demonstrated that an oscillating θ -angle can lead to underproduction of ${}^4\text{He}$ during big bang nucleosynthesis (BBN).

For our purposes it is the θ -dependence of nuclear binding energy that is of primary interest. Once nuclear binding energies become time dependent via (2) we can expect a periodic variation in the nuclear decay rates and hence the possibility to explain the reported decay anomalies.

In particular, we will demonstrate that tritium represents a particularly opportune target to search for these effects. This established, we will then use existing ${}^3\text{H}$ data to search for the corresponding axion-induced signal. Finding no statistically significant excess, we can then exclude axion decay constants below $9.4 \times 10^{12} - 1.8 \times 10^{10}$ GeV (95% confidence level) for masses in the $1.7 \times 10^{-23} - 8.7 \times 10^{-21}$ eV range. Conclusions and discussion are presented in closing.

II. θ -DEPENDENCE OF NUCLEAR DECAY RATES

Our underlying quantity of interest is the fractional change in the beta decay rate $\Gamma(\theta)$ as a function of θ ,

$$I_0(\theta) \equiv \frac{\Gamma(\theta) - \Gamma(0)}{\Gamma(0)}, \quad \Gamma(\theta) = \int_{m_e}^{E_{\text{max}} + \delta E(\theta)} dE_e \frac{d\Gamma}{dE_e}, \quad (3)$$

where $E_{\text{max}} = (M_i^2 + m_e^2 - (M_f + m_\nu)^2)/2M_i$, with initial and final state masses $M_{i/f}$, neutrino and electron masses $m_{\nu/e}$, E_e the energy of the emitted electron, and $\delta E(\theta)$ an additional θ -dependent contribution.

In principle this θ -dependence can also enter in other ways, for example modification of the underlying nuclear couplings $g_{A/V}$. However, in line with the results of Ref. [32] it is reasonable to assume the leading-order contributions arise specifically from modifications of the phase space. This point is also specifically analyzed in the Supplementary Material [33], in support of this conclusion.

By approximating nuclear beta decay in terms of free neutron decay, it has also been argued in Ref. [2] that for small perturbations to the decay rate, decays with smaller $Q \equiv M_i - M_f - m_e$ resulted in a larger fractional change in the beta decay rate.

However, from the beta decay rates given in Refs. [34–36] we can in any case evaluate this integral numerically for small $\delta E(\theta)$ without the free-neutron approximation, finding for ${}^{187}\text{Re}$ ($Q \simeq 2.6$ keV) and ${}^3\text{H}$ ($Q \simeq 18.6$ keV) that

$$I_0(\theta)|_{{}^{187}\text{Re}} \simeq 1.16 \left(\frac{\delta E(\theta)}{\text{keV}} \right), \quad I_0(\theta)|_{{}^3\text{H}} \simeq 0.18 \left(\frac{\delta E(\theta)}{\text{keV}} \right). \quad (4)$$

This result is particularly fortuitous in that if we wish to search for this effect, high quality datasets for ${}^3\text{H}$ already exist. The use of ${}^3\text{H}$ decay as a window to new physics more generally has also recently been explored in Refs. [37–39]. Furthermore, in addition to being more sensitive to the effect of a time-varying θ angle, the θ -dependence of the properties of lighter nuclei such as ${}^3\text{H}$ may also be comparatively easier to discern.

Indeed, in Ref. [32] the dependence of the nuclear binding energy upon θ has already been calculated for light nuclei. It was estimated that for three- and four-nucleon systems the n -nucleon binding energy \bar{B}_n satisfies

$$\bar{B}_n(\theta)^{1/4} - \bar{B}_2(\theta)^{1/4} = \bar{B}_n(0)^{1/4} - \bar{B}_2(0)^{1/4}. \quad (5)$$

Here the bar indicates an average over states which become degenerate in the limit that the approximate Wigner $SU(4)$ symmetry of low-energy nuclear physics becomes exact. As noted in Ref. [32] this agrees with the results calculated numerically in Ref. [40].

The averaged 2-nucleon binding energy $\bar{B}_2(\theta)$ can be found by averaging over the physical deuteron and the spin singlet (dineutron and diproton) channel, with θ dependence parametrized via

$$B_2(\theta) = \left(B_2(0) + \sum_{i=1}^3 c_i (1 - \cos \theta)^i \right) \text{ MeV}, \quad (6)$$

where $B_2(0) = 2.22, -0.072, -0.787$ MeV for the deuteron, dineutron and diproton respectively, and the c_i are numerical coefficients given in Ref. [32] (herein we choose the more conservative parameter set II, assuming isospin conservation).

Since this only gives the (averaged) $\bar{B}_3(\theta)$, rather than the individual initial and final state binding energies $B(\theta)_{i/f}$, we assume in the following that for small θ

$$B(\theta)_{i/f} \simeq B(0)_{i/f} \frac{\bar{B}_3(\theta)}{\bar{B}_3(0)}, \quad (7)$$

which is equivalent to assuming that for small θ the individual binding energies scale in proportion to their average.

Knowing the θ -dependence of the binding energy, we can then calculate $\delta M_{i/f}$, the θ -dependent part of $M_{i/f}$, via

$$M_{i/f} = \sum_N m_N(\theta) - B(\theta)_{i/f}, \quad (8)$$

where the sum runs over nucleons, and the neutron/proton mass difference is given by

$$m_n - m_p \simeq -0.58 \text{ MeV} + 4c_5 B_0 \frac{m_\pi^2}{m_\pi^2(\theta)} (m_u - m_d), \quad (9)$$

where $c_5 = (-0.074 \pm 0.006) \text{ GeV}^{-1}$ is a low-energy constant, $B_0 = m_\pi^2/(m_u + m_d)$, and we assume two degenerate quark flavors, following Ref. [32,41].

Expanding about $\theta = 0$ we find

$$\delta E(\theta) \simeq \delta M_i - \delta M_f, \quad (10)$$

which in turn can be calculated via Eqs. (7) and (9) since we know the θ -dependent parts of $(m_n - m_p)$ and $(B_i - B_f)$. In turn this allows us to calculate $I_0(\theta)$.

However, care is required here. In a universe where this mechanism is active the measured values of physical quantities such as decay rates are actually not those at $\theta = 0$, but are instead those at $\langle \theta^2 \rangle$ [since binding energies are $\mathcal{O}(\theta^2)$ at leading order]. Therefore when dealing with experimental data, rather than comparing $\Gamma(\theta)$ to $\Gamma(0)$, we should instead consider

$$I(\theta) \equiv \frac{\Gamma(\theta) - \langle \Gamma \rangle}{\langle \Gamma \rangle} = \frac{\Gamma(\theta)}{\Gamma(0)} \left(\frac{\Gamma(0)}{\langle \Gamma \rangle} \right) - 1, \quad (11)$$

where $\langle \Gamma \rangle$ is the average value of $\Gamma(\theta)$. Using the known $I_0(\theta) = \Gamma(\theta)/\Gamma(0) - 1$, we find after some calculation

$$I(\theta) \simeq 1.3 \times 10^{-5} \left(\frac{\rho_{\text{DM}}}{0.45 \text{ GeV/cm}^3} \right) \left(\frac{10^{-21} \text{ eV}}{m_a} \right)^2 \times \left(\frac{10^{13} \text{ GeV}}{f_a} \right)^2 \cos(2(\omega t + \vec{p} \cdot \vec{x} + \phi)), \quad (12)$$

which oscillates about zero, as expected, leading to alternating excesses and deficits in the number of nuclear decays per unit time. We can also see that the corresponding shift in the decay energy,

$$|\delta E(\theta)| \simeq 3.5 \times 10^{-2} \text{ eV} \left(\frac{\rho_{\text{DM}}}{0.45 \text{ GeV/cm}^3} \right) \left(\frac{10^{-21} \text{ eV}}{m_a} \right)^2 \times \left(\frac{10^{13} \text{ GeV}}{f_a} \right)^2 \quad (13)$$

is also a small correction in regions of the axion parameter space we exclude, justifying our assumptions that $\delta E \ll Q$. Similarly, $\theta \ll 1$ in these regions.

III. DATA ANALYSIS AND RESULTS

These points established, we can now search for evidence of this effect in ${}^3\text{H}$. We make use of a dataset, shown in Fig. 1, provided by the European Commission's Joint Research Centre (JRC) at the Directorate for Nuclear Safety and Security in Belgium, which spans approximately 12 years of liquid scintillation counter observations of the decay of an $\mathcal{O}(\text{microcurie})$ ${}^3\text{H}$ source [9].

It is worth emphasizing that the statistical analysis of nuclear decay anomalies has been subject to a number of

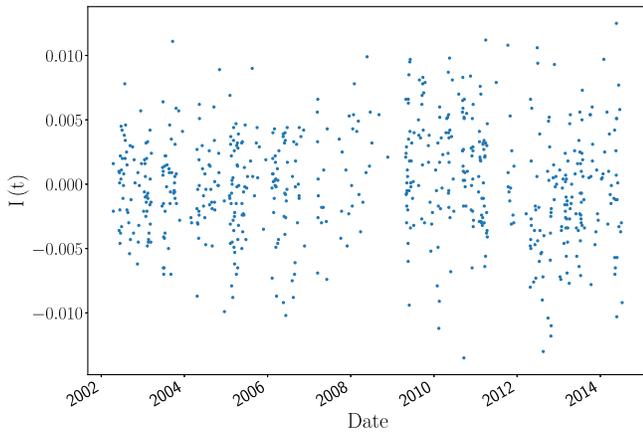


FIG. 1. Time-series data from Ref. [9], showing the fractional change in the ${}^3\text{H}$ beta decay rate, $I(t) = (\dot{N} - \langle \dot{N} \rangle) / \langle \dot{N} \rangle$. Here \dot{N} is the measured value of decays per second, whilst $\langle \dot{N} \rangle$ is its expected value due to the exponential decay law.

treatments [42], with no overall consensus on which approach is optimal. This being the case we broadly follow the approach of Refs. [29,30,43], where searches for oscillatory signals in time-series data were also used to place limits on ultralight axion DM.

Since the data points are unevenly spaced in time we will estimate their power spectrum using the least-squares spectral analysis (LSSA) method to construct periodograms [44,45]. We compute the power spectrum using the `astropy.timeseries.LombScargle` class provided by the Python `astropy.timeseries` package [46], evaluated at a set of 8113 evenly spaced trial frequencies.

As the lowest 15 trial frequencies appear to show evidence of uncontrolled systematic effects, possibly long-term drift of the experimental apparatus, we exclude them from our analysis. The largest frequency in the remaining data, 4.2×10^{-6} Hz, corresponds to a period of ~ 2.8 days, whilst the smallest, 8.0×10^{-9} Hz, corresponds to a period of ~ 4.0 years. The resulting periodogram is then given by the blue curve in Fig. 2.

Under the null hypothesis that the dataset contains no axion-induced signal, the time-series datapoints should follow a Gaussian distribution about $I = 0$. To place limits on the corresponding power spectrum we therefore perform Monte Carlo (MC) simulations by generating $N = 50,000$ time-series MC datasets, with the same time spacing as the original data. The MC data points themselves are drawn from a zero-mean Gaussian distribution, with width set by the standard deviation of the original dataset.

For each MC dataset we can calculate a corresponding periodogram, and then use the statistics of these periodograms to construct the cumulative distribution function (CDF) for the power at each frequency. From these CDFs we then determine the false positive (or false alarm) power at 95% confidence level for each frequency, as shown in

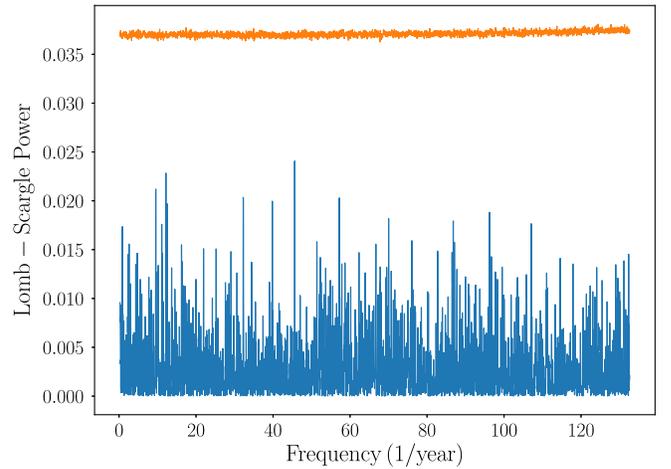


FIG. 2. Periodogram corresponding to the dataset shown in Fig. 1 (blue), along with the MC-derived 95% confidence level threshold (orange). As can be easily seen, the data are compatible with the null hypothesis.

Fig. 2 in orange. Here and in the following we account for the “look elsewhere effect” by defining these limits with respect to the global trials factor $p_{\text{global}} = 1 - (1 - p_{\text{local}})^{N_f}$, where p_{local} is the corresponding local p -value and N_f the number of trial frequencies. As can be seen, nowhere does the original dataset exceed the 95% confidence level threshold, indicating the data are compatible with the null hypothesis.

To determine the corresponding limit on the axion parameters we follow a similar approach. For each trial frequency we construct $N = 50,000$ MC datasets containing Gaussian background and injected axion-derived signals and calculate their periodograms. The injected signals are of fixed amplitude and frequency, with the unknown axion phase ϕ drawn from a uniform distribution. As the injected signals are constructed in time series form, we directly match the axion velocity to the lab-frame DM velocity, incorporating modulation effects due to the Earth’s passage through the DM halo [47,48]. For a given choice of parameters, we can then construct the corresponding CDF for the power at each trial frequency. With the mass fixed by the frequency under consideration, the threshold value of f_a can then be determined following a standard frequentist approach, well illustrated in Ref. [49]. Having determined the threshold value of f_a within this framework, we then subsequently account for the “stochastic vs deterministic” correction factor occurring in the regime where the measurement time is much less than axion-field coherence time, following Ref. [49].

Specifically, from the background-only CDF we first find the false positive threshold power at a desired confidence level. From the background plus signal CDF we can also find the false negative threshold power, for a given choice of f_a . The threshold value of f_a is then determined by the condition that the false positive threshold from the

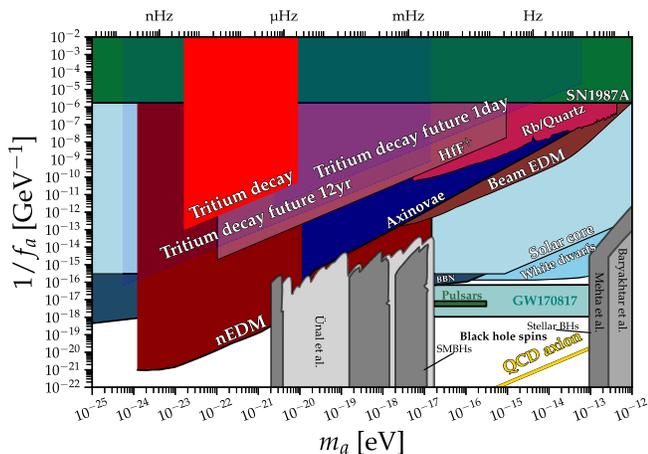


FIG. 3. Axion DM constraint from the nonobservation of periodic variations in ${}^3\text{H}$ decay data. We exclude f_a below $9.4 \times 10^{12} - 1.8 \times 10^{10}$ GeV (95% confidence level) for masses in the $1.7 \times 10^{-23} - 8.7 \times 10^{-21}$ eV range. Also shown are constraints from oscillating nEDM searches [27–30], BBN [31], the spectroscopy of radio-frequency atomic transitions [50], pulsars, gravitational waves and black hole superradiance [51–55], solar/white dwarf observations [56,57], so-called “axinovae” [58], and the parameter space occupied by canonical QCD axion models (yellow). Figure producing using the AxionLimits code [59].

background-only CDF coincides with the false negative threshold from the background plus signal CDF, at the desired confidence level. Equivalently we can say that the threshold value of f_a occurs when the false positive rate α is equal to the false negative rate $1 - \beta$, where $CL = 1 - \alpha$. The resulting exclusion curve is given in Fig. 3.

We simulate future experimental possibilities, via two fiducial cases also shown in Fig. 3. The first is a long-term experiment designed to search lower frequencies, with one measurement per hour over 12 years. The second is a short-term experiment to search higher frequencies, with approximately one measurement per second over one day. Both schemes increase the initial quantity of tritium by a factor of 100 relative to the JRC dataset, up to a presumed limit imposed by detector pileup [60]. In the first case we can exclude axion decay constants up to $f_a < 1.5 \times 10^{16}$ GeV (95% confidence level) and cover masses in the $5.4 \times 10^{-25} - 1.4 \times 10^{-17}$ eV range, while in the latter we can exclude axion decay constants up to $f_a < 4.5 \times 10^{11}$ GeV (95% confidence level) and cover masses in the $2.4 \times 10^{-21} - 6.3 \times 10^{-14}$ eV range.

IV. CONCLUSIONS/DISCUSSION

We have explored purported variations in nuclear decay rates, providing a novel and well-motivated explanatory mechanism within the framework of axion physics. The oscillating QCD θ -angle created by the misalignment

mechanism induces a time-varying nuclear binding energy, which then leads to the time-dependence of nuclear decays.

Analyzing 12 years of ${}^3\text{H}$ decay data we find no corresponding statistically significant excess, and therefore exclude axion decay constants below $9.4 \times 10^{12} - 1.8 \times 10^{10}$ GeV (95% confidence level) for masses in the $1.7 \times 10^{-23} - 8.7 \times 10^{-21}$ eV range.

One advantage of this approach is that we can probe regions of axion parameter space which normally are only accessible experimentally via much more sophisticated methods, such as oscillating nEDM searches [27–30]. In contrast, the data we have analyzed here were taken using a single $\mathcal{O}(\text{microcurie})$ tritium source and a commercially available laboratory liquid scintillation counter. Of course, there are also astrophysical and cosmological bounds on the mass of axion dark matter, such as $m_a > 3 \times 10^{-19}$ eV (99% confidence level), coming from observations of ultra-faint dwarf galaxies [61] and $m_a < 2 \times 10^{-20}$ eV (95% confidence level) bounds from the Lyman-alpha forest [62]. However, our work provides a new kind of laboratory-based scheme to search for QCD axion dark matter, which is complementary to such constraints without relying on cosmological or astrophysical assumptions and modeling.

However, relying on nuclear decays in this way also presents certain challenges. In particular, issues such as detector pileup may ultimately constrain sensitivity more so than in other experimental approaches.

Another overall consideration here is the validity of this and other approaches in searching for QCD axion-derived phenomena far away from the parameter space where these effects are expected to occur; the so-called QCD axion band. It is first of all important to note that (admittedly nonminimal) models of the QCD axion do exist in the parameter space we have probed here, as given in Refs. [56,63–65]. Furthermore, we can also understand our efforts here as being a necessary step to ultimately reach the sensitivity to probe standard QCD axion models via methods such as these.

There are several avenues by which these findings could be improved. On the theoretical side, more accurate estimation of the dependence of nuclear binding energies on θ would be of use. Experimentally, dedicated nuclear decay experiments which are optimised to search for this particular effect should also yield stronger constraints, and may be able to probe previously unexplored regions of the axion parameter space. Analysis of other preexisting nuclear decay datasets may also similarly bear fruit.

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