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Letter

New tests of quantum mechanics with unstable nuclei

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ABSTRACT

We propose and analyze two new tests of quantum mechanics that would probe the time evolution of unstable nuclei. The first is a search for a time-dependent nuclear magnetic moment in radioisotopes over several half-lives. The second is a novel application of the Mössbauer effect to search for a time-dependent nuclear transition frequency in a radioisotope over several half-lives. The complexity of non-perturbative quantum chromodynamics (QCD) makes unstable nuclei promising candidates for the observation of deviations from quantum mechanics.

1. Introduction and background

This year marks one century since Erwin Schrödinger postulated his famous equation; it is the basis for the highly successful theory of quantum mechanics that has passed all experimental tests to date. The Schrödinger equation is time-symmetric, in contrast to dissipative systems where there is an "arrow of time" [1,2]. The decay of an unstable quantum system is described using a density matrix [3], and alternatively by quantum trajectories where state collapse is inserted randomly [4–5]. This framework is a working approach that agrees with all experiments to date, such as quantum jumps in the spontaneous emission of atoms [6]. Despite this success, there is a general agreement that the current quantum theory is incomplete since it does not include interactions with the environment in a fundamental way. The key question is whether there are any new experimental tests that could violate the current quantum theory, requiring its revision.

This tension has led to recent theoretical models that extend quantum mechanics and naturally incorporate interaction with the environment [7–9]. In an earlier paper, we proposed that an atomic clock of a radioisotope could exhibit an "aging effect" where the clock frequency varies over several half-lives as it becomes entangled with the environment [10]. In this paper, we propose to search for new effects beyond standard quantum mechanics in unstable nuclei with half-lives ranging from several days to weeks. The first is to measure the hyperfine splitting over time with a radioactive ion microwave clock. The second is to search for time-dependent nuclear transition frequencies using a novel variation of Mössbauer spectroscopy [11].

2. Nuclear magnetic moments

Precision measurements of nuclear magnetic moments are mostly based on microwave-optical double resonance spectroscopy of atomic hyperfine splitting which is proportional to the nuclear magnetic moment [12]. For example, the current atomic frequency standard is based on the ground-state hyperfine splitting of the cesium atom in a laser-cooled atomic fountain [13]. Single trapped ions are used for precision measurements of the nuclear magnetic moment, both in Penning traps [14–15] and radio-frequency Paul traps [16–17]. The accuracy that is reached is around 10^{-13}, requiring several days of averaging, which far exceeds ab initio calculations, limited to an accuracy of 1–10 % due to the computational challenge of non-perturbative QCD. To search for a time-dependent nuclear magnetic moment in unstable nuclei at the above accuracy requires half-lives of several weeks and convenient transitions for laser cooling and detection of the trapped ion.

In an earlier paper, we presented a theoretical model for "atom aging" which introduced a nonlinearity in quantum mechanics that accounted for quantum entanglement of the unstable system with the environment [10]. In that work, we proposed optical clock transitions in trapped Lu-177 and Ra-223 ions. In the current paper, we focus on microwave transitions rather than optical transitions and provide realistic cases. Probing the nucleus is more likely to lead to discovery than electronic transitions due to the relatively poor understanding of non-perturbative QCD; microwave clocks with trapped ions are much simpler than optical clocks; Production of the above radioisotopes at high specific activity can be accomplished with new methods of isotope separation; precision measurements on trapped ions require several days

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of averaging, so the proposed radioisotopes must have sufficiently long half-lives for this purpose.

In the context of the theoretical framework from ref. 10, to observe any effect beyond quantum mechanics it is necessary to polarize the nuclear spin in a fixed external axis, denoted by z, otherwise the nonlinear quantum effect will average out. As explained by Surjeet Rajendran (private communication): one cannot have a polarization mechanism where one "measures" the spin and get spin up with 50 % probability and spin down with 50 % probability and only keep the spins that are spin up. This is because the process of measurement creates both outcomes and the apparatus are entangled to these outcomes. In linear quantum mechanics, these different outcomes cannot influence each other because of decoherence, but in nonlinear quantum mechanics they do. We note that nuclear polarization is needed in the laboratory frame, not affected by Earth's rotation.

One can estimate the frequency shift as follows (Surjeet Rajendran, private communication): The usual hyperfine splitting is proportional to $\mu_N\ \mu_e/A^3$ where μ_N is nuclear magnetic moment, μ_e is the electron magnetic moment and A is the Bohr radius.

With the non-linear effect, there will be a background magnetic field $\mu_N\ /L^3$ where L is the size of the ion's center of mass wavefunction in the trap, around 10 nm. The shift is $\xi\ \mu_N\ \mu_e\ /L^3$, where ξ is the unknown parameter. If a shift is measured at a level of 10^{-13}, then $\xi \sim 10^{-7}$. A null measurement would constrain theory at that level of accuracy.

More generally, observation of a time variation of the nuclear magnetic moment for unstable nuclei would be a discovery of new physics beyond quantum mechanics, *independent of any theoretical framework*. Such a test has not yet been performed.

There are numerous radioisotopes that could be used for such tests, starting from Be-7 with a half-life of 53.22 days. This light ion was indeed trapped and laser-cooled at an accelerator lab, but the measurement of its hyperfine splitting was done rapidly before the ions were released [18]. Radioactive barium isotopes (Ba-131 and Ba-133) were confined in an RF Paul trap and their hyperfine splitting was measured, but the ions were not laser-cooled, they were not tightly localized, and the precision was not very high [19]. Other promising isotopes listed here with their half-lives include Sr-89 (50.53 days), Sr-85 (64.85 days), Ca-45 (162.6 days), Yb-169 (32 days), and Ra-223 (11.43 days). These radioisotopes range from light to heavy, and emit beta, gamma, and alpha particles in their decay. Single ion clocks have already been demonstrated in Sr, Ca, and Yb, and Ra, so they are fully amenable to such measurements [20–23].

A simplified Grotrian diagram for $Sr\text{-}89^+$ is shown in Fig. 1.

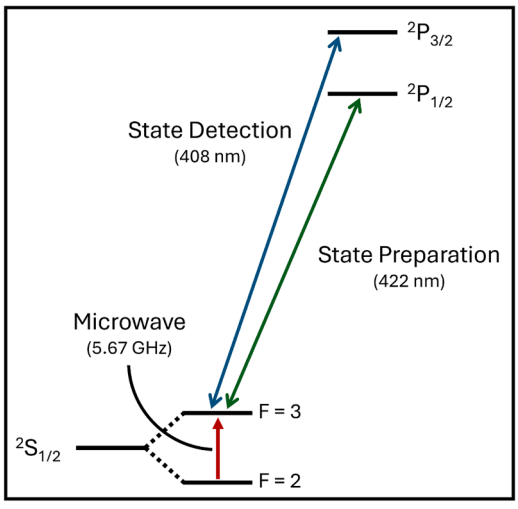

Fig. 1. A simplified Grotrian diagram of Sr+, showing the laser for state preparation near 422 nm, the laser for state detection near 408 nm, and the hyperfine splitting of the ground state of 5.67 GHz.

The ground state $F = 2$ to $F = 3$ hyperfine splitting is 5.67 GHz [24], which can be measured using microwave-optical double resonance spectroscopy. The ion's state is first prepared by optically pumping to the $F = 2$ state with a laser near 422 nm tuned to the upper $P_{1/2}$ state. The microwave drive is applied as a Ramsey pulse sequence, and the upper $F = 3$ hyperfine state is detected with a laser near 408 nm as a cycling transition with the upper $P_{3/2}$ state. To load a Sr-89 ion in an RF Paul trap (or multiple ions in a linear trap), neutral Sr-89 can be vaporized near the trap, and photo-ionized with two lasers near 461 nm and 405 nm that are focused to the center of the trap [25].

One challenge is that to efficiently load ions into a trap, high enrichment of the desired isotope is necessary. In the above cases, the stable precursors are the same elements, precluding the use of radiochemistry. Sr-89 is produced by neutron irradiation of stable enriched Sr-88, Sr-85 from stable Sr-84, Ca-45 from stable Ca-44, and Yb-169 from stable Yb-168. We have developed efficient methods of isotope separation that can meet this goal for the above cases [26–27]. It is worth noting that many radioisotopes can also be produced by combining resonant laser ionization with a magnetic dipole separator like at the MEDICIS facility at CERN [28].

3. Nuclear transitions

An alternative to searching for a time variation of nuclear magnetic moments in radioisotopes is to search for a time-variation in the transition frequencies in unstable nuclei. Direct excitation with a laser is generally not possible due to nuclear excitations being in the gamma range of the electromagnetic spectrum. There is one exception with Th-229 which has a transition in the vacuum ultra-violet part of the spectrum near 148 nm and is the basis for a nuclear clock [29–30]. Unfortunately, the half-life of Th-229 is almost 8000 years, so while a search for a time-dependent clock shift is worthwhile, this isotope is not amenable to a search for time-dependence over several half-lives.

We propose, instead, to search for a time-dependent nuclear transition using a novel variation of Mössbauer spectroscopy [11]. The Mössbauer effect, discovered in 1958 by Rudolf Mössbauer, is that recoilless emission and absorption of nuclear gamma rays can occur in solid crystals under the right conditions. The emitter is a radioisotope that decays into the absorber, which is a stable isotope. A commonly used isotope pair is Co-57/Fe-57, where the cobalt isotope has a half-life of around 272 days, and decays by electron-conversion to the iron isotope. The last emission in the decay of Co-57 is a gamma photon near 14.4 keV, which has a lifetime of about 100 ns. The recoilless fraction, f, depends on the gamma energy, temperature, mass of the emitting atom, and Debye temperature of the crystal. The Co-57/Fe-57 Mossbauer pair is very convenient since they can be maintained at room temperature. Mössbauer spectroscopy is performed by mounting the emitter on a translation stage that can move at a controlled velocity to induce a Doppler shift on the emitted gamma, as illustrated in Fig. 2. The resonance is measured by varying the emitter velocity in the range of several cm/s, as shown schematically in Fig. 3.

Over the years since its discovery, the Mössbauer effect has mainly been used to probe solid state effects. A landmark experiment in 1960 used Mössbauer spectroscopy to directly measure the gravitational redshift [31]. In that work, known as the Pound-Rebka experiment, the emitter (Co-57) and the absorber (Fe-57) were separated vertically by 22.5 m. The authors found that the Doppler shift needed to compensate for a frequency shift was exactly as predicted by Einstein's general theory of relativity.

We propose here a variation of Mössbauer spectroscopy by using *two radioisotopes*, one as emitter and the other as absorber. Each of the two isotopes is characterized by its own half-life, and by varying the age of one versus the other, it would be possible to search for a time variation of a nuclear transition frequency. Instead of a Mössbauer pair being separated in space as in the Pound-Rebka experiment, *they would be separated in time*. This test would be outside any current theoretical

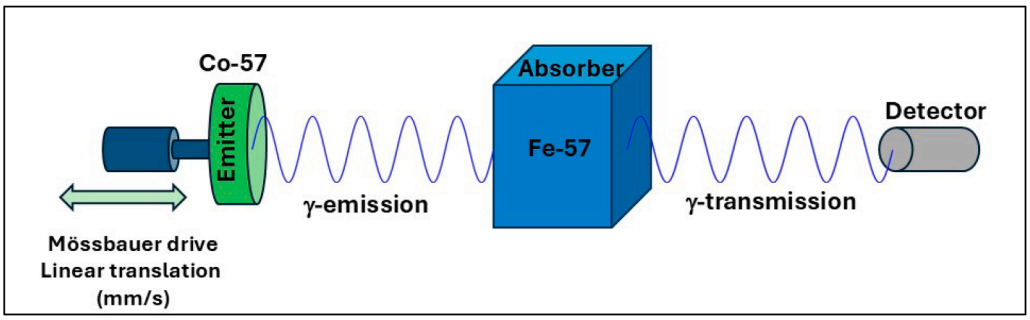

Fig. 2. Schematic diagram of a typical Mössbauer spectroscopy experiment. In the case shown for the Co-57/Fe-57 pair, the emitted photon is at an energy of 14.4 keV.

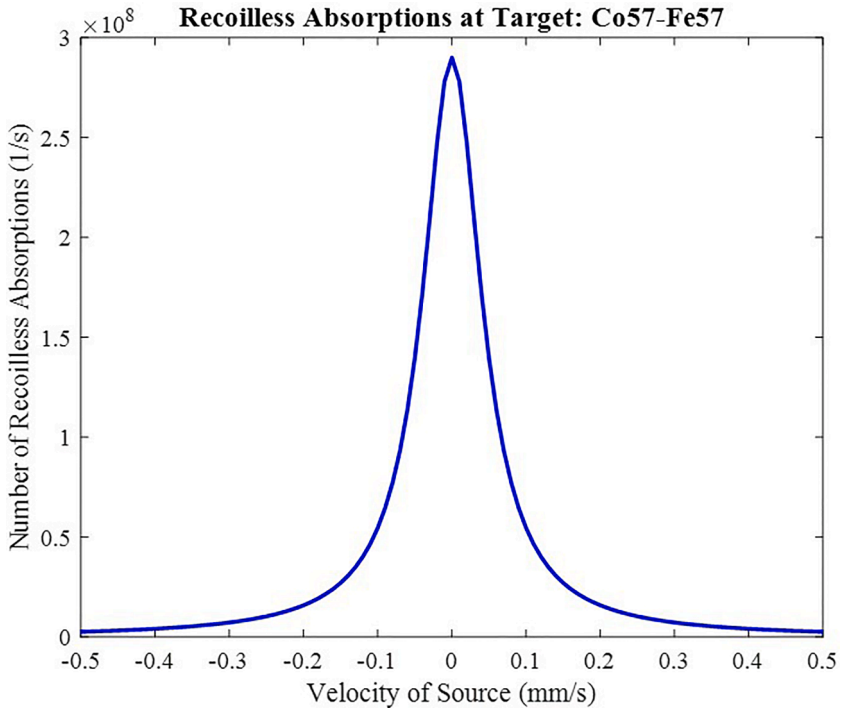

Fig. 3. Schematic plot of a resonance absorption scan in the Pound-Rebka experiment. The scan velocity is shown in the horizontal axis (in mm/s) and the vertical axis has the number of recoilless absorptions.

model, such as our previous atom aging paper.

The question is whether one can find an appropriate Mössbauer pair which satisfies many constraints. The emitter half-life should ideally be long (months), yet the last emitted gamma should have a short half-life so that the Mössbauer resonance is not too narrow or too broad (typically 0.1–10 nanoseconds). The gamma energy should be <150 keV, considered to be an upper limit for recoilless emission and absorption. The absorber half-life should also be short enough to measure over several half-lives, yet longer than the time needed to produce it in a nuclear reactor. The radioisotope pair must be produced with high efficiency in a nuclear reactor to reach the necessary level of activity, and they must be separated from each other to avoid interferences. The gamma detectors must be able to resolve the desired line, rejecting other gamma emissions. Finally, the radioisotope pair must be imbedded in crystals with a high Debye temperature to achieve a good fraction of recoilless emission and absorption.

We have conducted a detailed search and found only one radioisotope pair that satisfies all the above criteria. It is Lutetium-177 (Lu-177), used for cancer therapy, and its nuclear isomer state Lutetium-177 m (Lu-177 m). Lu-177 m has a half-life of 160 days, and decays by isomeric transition to Lu-177 in 22.7 % of the time (the rest decays to stable Hf-177). The last gamma emission in the decay chain to Lu-177 has an energy of 122 keV and a half-life of around 0.1 nanoseconds. Lu-177 has a half-life of 6.64 days, as shown in the decay diagram in Fig. 4.

Lu-177 can be produced by neutron irradiation of stable Yb-176 or by neutron irradiation of stable Lu-176, the latter has a neutron cross section of over 2000 barns which would enable production of very high activity material (hundreds of Curies or more). Production of Lu-177 m would also be done by neutron irradiation of Lu-176, where the ratio of Lu-177 to Lu-177 m is about 4:1 [32].

Efficient separation of Lu-177 and Lu-177 m from Lu-176 can be accomplished with our methods [25], and the experimental realization is in progress in our group, motivated by the need for Lu-177 in targeted cancer therapy.

These isotopes (or their stable precursors) can be sintered as oxides with another host, such as aluminum oxide or yttrium aluminum oxide (YAG) to form crystals with high Debye temperatures. Detection of gammas at 122 keV can be done with very high efficiency and selectivity with a new generation of detectors used for single-photon medical imaging. Specifically, CdZnTe (CZT) detectors have demonstrated

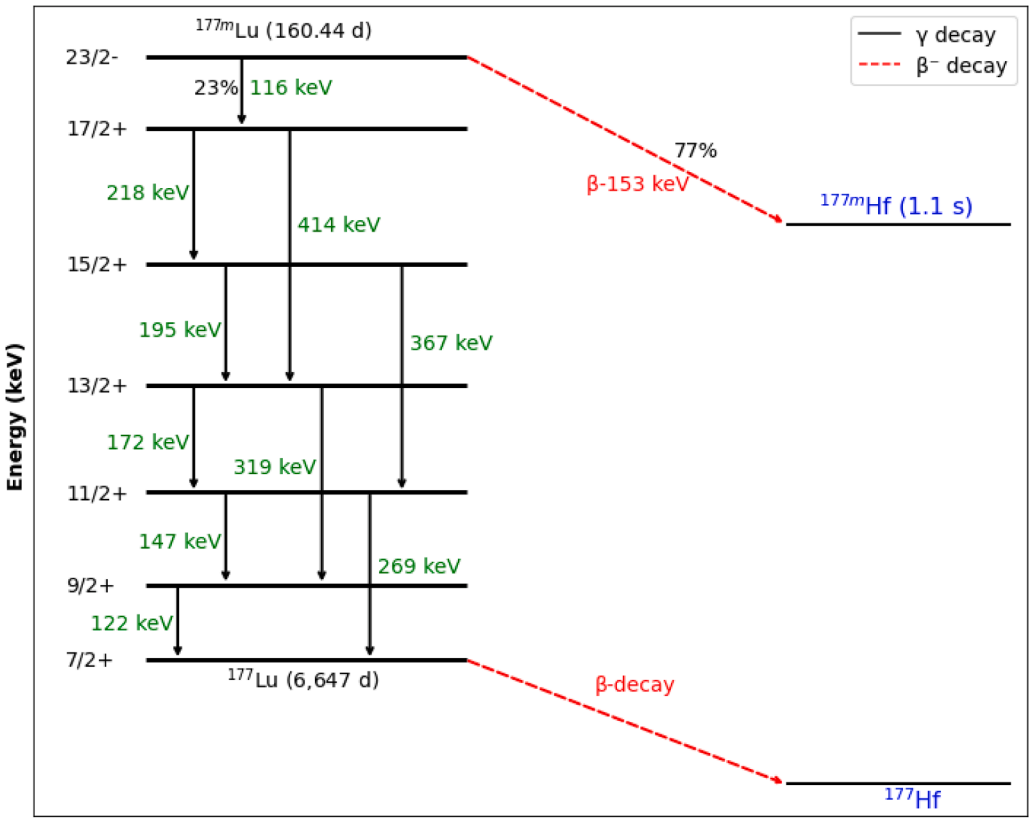

Fig. 4. Decay diagram of Lu-177 m and Lu-177.

ultrahigh energy resolution of about 1 % at 122 keV [33].

We have calculated the recoilless fraction for this Mössbauer pair at room temperature, assuming a Debye temperature of 980 K for aluminum oxide, and it is 0.38. At liquid nitrogen temperature of 77 K, the fraction is 0.78. Both cases should be adequate for Mössbauer spectroscopy. The required velocity scan is in the range of cm/s, and a

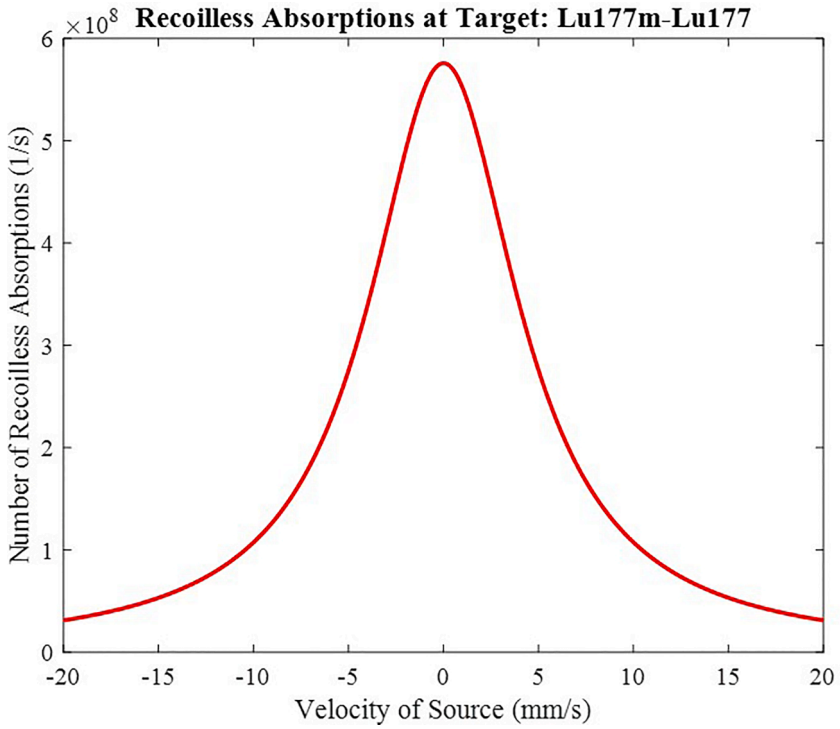

Fig. 5. Schematic plot of a resonance absorption scan for the Lu-177m/Lu-177 Mössbauer pair. The scan velocity is shown in the horizontal axis (in mm/s) and the vertical axis has the number of recoilless absorptions.

schematic plot of an expected resonance is shown in Fig. 5. This assumes a 5 Ci Lu-177 m source and an 8 cm (0.1 steradian area) diameter target holding 100 Curies of Lu-177. These numbers are very realistic based on irradiation at the University of Texas at Austin Research Reactor (William Charlton, private communication). As the Lu-177 m and Lu-177 decay to Hf-177, there could be a systematic effect in the absorption scan due to the change of the lattice. This can be tested by varying the age of the Lu-177 m, and by changing the concentration of Lu-177 in the host lattice.

In summary, this novel version of Mössbauer spectroscopy looks feasible from the technical standpoint and could provide a new window into the nucleus.

4. Conclusions

We propose here two new experimental tests of quantum mechanics that have never been proposed or performed. Both involve the atomic nucleus which is still poorly understood at the level of precision measurement due to the complexity of non-perturbative QCD. These tests address a central question in quantum mechanics about the role of time in unstable systems and hold the prospect of discovery of new physics.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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