EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time of Flight Committee

Precision measurement of the half-life of ^{110}Sn in large and small lattice environments

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Abstract

We propose to undertake high-precision measurements of the half-life of electron-capturing nuclear decay rate of ^{110}Sn under compression by implanting the ions in a small lattice such as palladium (Pd) and a large lattice such as lead (Pb). Such studies have implications in many areas ranging from astrophysics to geophysics as well as for the measurement of high precision beta transitions essential for the fundamental tests of the standard model. At present, very little data is available on the change of electron capture decay rate under compression and the available data seems to indicate that the observed increase of the electron capture decay rate under compression is much greater than the predictions of the state-of-the-art density functional calculations as obtained from TB-LMTO or WIEN2K codes. The proposed experiment should generate very high precision data thus clarifying the experimental situation. High purity, intense and energetic (2.2 MeV/A) ^{110}Sn beam available from HIE-ISOLDE provides a unique opportunity for clean implantation to perform such measurements.

Requested shifts: 5 shifts (each shift is 8 hours long)

1 Motivation:

Radioactive decays are generally known to be independent of external pressure, temperature and chemical environment [1]. Among the various types of radioactive decays, electron capturing nuclear β-decay rate [2] is known to be slightly affected by the external environment [3-9]. The electron capture nuclear decay rate is proportional to the electron density at the nucleus$[|\psi(0)|^2]$, where $\psi(0)$ is the electronic wave function at the nucleus [2]. Since the atomic valence electron configuration could be modified by the surrounding environment and that would slightly affect the electron density at the atomic nucleus $\psi(0)$, the electron capture nuclear decay rate is susceptible to the surrounding environment. ^{7}Be is the lightest nucleus that decays by electron capture and its decay rate is expected to be most susceptible to the surrounding environment because its electronic configuration is $1s^2 2s^2$ and the valence 2s electrons contribute ≈3.2% to the total electron capture decay rate. The decay rate of ^{7}Be in different media has been extensively studied [1, 3-9]. It was found that the electron affinity of the surrounding medium could affect the decay rate of ^{7}Be by measurable amount and density functional calculations provided reasonable agreement with the experimental results for such cases [5, 9]. There was a debate regarding the importance of the conduction electrons of metal (Debye screening) to modify the radioactive decay rate in metals versus insulators [10]. However, the non-observation of any observable temperature dependence of radioactive decay rates essentially ruled out such explanations [11,12]. Moreover, density functional calculations predict negligible effect of the conduction electrons on the electron density at the nucleus.

On the other hand, the change of electron capture nuclear decay rate under compression is not at all well studied. However, it is important to understand the variation of electron capture nuclear decay rate under compression, because of its various astrophysical and geophysical significances. It is believed that the heavy element creation takes place during the merger of the neutron stars and core collapse of the massive stars [13,14] and the electron capture by the nuclei plays an important role in these processes. The electron capture nuclear decay is important in heating up the earth's core where the pressure is very high (3.3 Mbar – 3.6 Mbar). A large amount of heat (about 8 TW power) could be produced [15] by the electron capture of ^{40}K in the core alone and this heat production is important in determining the thermal and tectonic evolution of the earth. Moreover any unexplained increase of electron capture nuclear decay rate under compression would have implications for high precision beta decay measurements [11] that are essential for the tests of standard model. So it is important to understand the change of electron capture nuclear decay rate under compression. However there is very little data available so far except for ^{7}Be. W. K. Hensley et al. [16] found that the decay rate of ^{7}Be increases by about 0.6% when ^{7}BeO lattice is compressed at a pressure of 27 GPa. Liu and Huh [17] applied high pressure on amorphous $^{7}Be(OH)_2$ gel and obtained similar results. On the other hand, density functional and Hartree-Fock calculations [18-21] predicted ~(0.1-0.2)% increase of the electron density at the ^{7}Be nucleus for the application of 27 GPa pressure on ^{7}BeO lattice. Recently, it was found [22] that the decay rate of ^{7}Be increases by (0.82±0.16)% when implanted in small Pd lattice versus large Pb lattice, whereas density functional calculations predict only an increase of about 0.2%.

In the cases of the compression of the larger many-electron radioactive (electron capturing) atoms such as ^{40}K, ^{109}In, ^{110}Sn etc., the increase of the electron density at the nucleus is generally expected to be very small at the compressional pressures achievable in the laboratory experiments, because only the valence orbitals could be compressed and the overlap of the valence orbital electrons (such as 4s, 5s etc.) at the nucleus is exceedingly small, compared to the overlap of the inner orbital electrons. K. K. M. Lee and G. Steinle-Neumann performed [18] WIEN2k density functional calculations [19] to calculate the increase of electron density at ^{40}K nucleus for the compression of $^{40}K_2O$ and found that the electron density at the ^{40}K nucleus should increase by < 0.01% even at a pressure of 50 GPa. One way to experimentally study the effect of compression on the electron capture nuclear decay rate could be to implant the relevant radioactive atoms/ions in the interstitial spaces of a small and large crystal lattice and look for any change of the decay rate. There is one set of experimental data showing [23] that the orbital electron capture nuclear decay rates of ^{109}In and ^{110}Sn increased substantially by (1.00±0.17)% and (0.48±0.25)% respectively when implanted in the smaller Au lattice compared to the larger Pb lattice. The electron affinity of Au (electron affinity=2.2 eV) is much larger than that of Pb (electron affinity=0.35 eV) [24]. However, the electron affinity of the surrounding medium should primarily affect the valence electrons of ^{110}Sn ions and hence its effect is expected to be negligible in this case. So, the observed relatively large increase of decay rate is probably due to the compressional effect in the smaller Au lattice. Our density functional calculations using WIEN2k code [19] indicate only about 0.1% increase of decay rate of ^{109}In in Au compared to that in Pb, even after considering

finite nuclear size and quantum electrodynamics effects. So the reported large increase of decay rate has remained unexplained so far.

Justifications for performing ^{110}Sn experiment at CERN HIE-ISOLDE and the choice of incident energy

Earlier study [23] of the decay rate of ^{110}Sn was done by implanting ^{110}Sn ions in Au and Pb catcher foils along with all other radioactive ions produced in the heavy ion reaction ^{20}Ne+^{93}Nb at E(^{20}Ne)= 80 MeV. The peak area of 280 keV γ-ray emitted by ^{110}In following electron capture by ^{110}Sn, was monitored with time to determine the half-life of ^{110}Sn in different catcher foils. However, the ratio of peak to valley for the 280 keV γ-ray line was of the order of 10 [23], because of the background coming from many other sources. The area of the peak was determined by subtracting out the background under the peak. The main uncertainties in the determination of the peak area came from the choice of points on the two sides of the peak to draw the background. At CERN HIE-ISOLDE, primarily ^{110}Sn ions are expected to be implanted in the catcher foils and 280 keV γ-ray line is expected to be the main peak in the spectrum. So the background under the 280 keV γ-ray is expected to be very low resulting in dramatically improved peak to valley ratio. Since ^{110}Sn is a relatively intense beam (10^5 ions/s) at HIE-ISOLDE, the peak to valley ratio on the order of 1000 is expected, thus significantly reducing the uncertainty in the determination of the peak areas and half-life measurement with better than 0.05% accuracy should be possible [25].

Another issue is that radioactive ions should be implanted in the bulk region of the catcher foil to study the effect of lattice compression. If we use 50-60 keV ^{110}Sn ions from CERN ISOLDE for the implantations on the palladium (Pd) and lead (Pb) foils, SRIM code predicts that the range of ^{110}Sn ions in Pd would be ~100Å with a straggling length of ~60Å and the corresponding range in Pb would be ~150Å with a straggling length of ~125Å. Considering the uncertainties in SRIM calculations at these low energies, the implantations would be very close to the surface of the catcher foils. Since there could be oxide layer, adsorption of water and other contaminants at the surface of the catcher foil, it is not a good idea to implant at the surface of the catcher foil, because the measurements could be affected by the surface effects. Another source of systematic error could be the possible diffusion of the implanted ions out of the catcher foils with time [26]. Hence, the implantation of ^{110}Sn ions very close to the surface of the catcher foils (within ~50 Å) could be a problem for the high precision measurement of the half-lives. Moreover, density functional calculations such as WIEN2k calculations [19] assume a periodic boundary condition and do not consider any surface boundary.

Hence, we propose to perform implantations using the available higher energy beam from HIE-ISOLDE. However, the incident energy of ^{110}Sn should be sufficiently below the Coulomb barrier of the ^{110}Sn and the nuclei of the catcher foils (Pd and Pb), so that no long-lived γ-emitting radioactive isotopes are produced due to the nuclear reactions. The relevant Coulomb barrier is around 4.5MeV/A (Laboratory frame) and the incident beam energy of ^{110}Sn ions should preferably be less than half of Coulomb barrier energy to significantly reduce the possibility of sub-barrier reactions. We propose to use ^{110}Sn ions (having energy 2.2 MeV/A), because it is relatively easy to obtain ions of this energy from HIE-ISOLDE. These energetic ions would be implanted at a depth of 10-15 μm inside the Pd and Pb catcher foils (thickness of foil =25 μm). So the implanted ions would be in the bulk region and would not be affected by any surface effect. Since no nuclear reaction would take place below Coulomb barrier, the catcher foils are expected to contain primarily the radioactive ^{110}Sn ions after the implantations. Hence we expect a clean implantation and ^{110}Sn ions are expected to experience compression at the interstitial spaces of the lattice. Since we propose to implant in small Pd lattice (lattice constant= 3.89Å) and large Pb lattice (lattice constant= 4.95Å) [24] and both Pb (electron affinity=0.35 eV) and Pd (electron affinity=0.54 eV) [24] have similarly low electron affinities, any observed increase of decay rate in Pd lattice could be unambiguously attributed to the higher compressional effect in Pd lattice.

2. Description of the experiment:

We propose to use a pure ^{110}Sn beam (having intensity ~10^5 ions/sec; energy =2.2 MeV/A from HIE-ISOLDE and implant the ions in Pd and Pb foils one by one. High energy (2.2 MeV/A) ^{110}Sn beam would penetrate 10-15μm inside Pd or Pb foils (thickness= 25 μm) and hence surface effects cannot be responsible for any observed change of decay rate. Moreover, we shall treat the surface of Pb foil chemically to remove any lead-oxide layer on it and then immediately put it in a high vacuum. The implantation of ^{110}Sn ions on each target would be for about 8 hours (half-

life of ^{110}Sn being ~4.2 hours). There would be essentially no nuclear reaction between ^{110}Sn and the nuclei of the catcher foils, because the relevant Coulomb barriers are much higher than the incident energy of ^{110}Sn ions. After implanting ^{110}Sn ions in a Pd or Pb foil for 8 hours, the implanted foil along with a standard ^{60}Co γ-ray source would be counted by placing it in front of a high efficiency HPGe detector or CLOVER detectors. The counting would be done in a low background room. The singles count rate of each HPGe detector would be kept below 10,000 counts per sec to enable good energy resolution (FWHM≈ 2 keV at 662 keV energy). The time keeping would be done by using a precision pulser. The composite γ-ray spectrum from the decay of ^{110}Sn ions (that emit 280 keV γ-ray photons following the electron capture nuclear decay of ^{110}Sn and subsequent production of excited ^{110}In) and ^{60}Co source (that emits 1173 keV and 1332.5 keV γ-ray photons) would be recorded in a data acquisition system. The ratio of the peak area of 280 keV γ-ray line from the decay of ^{110}Sn and the sum of the peak areas of ^{60}Co γ-ray lines would be monitored with time to cancel out the dead time effect of the data acquisition system. Simultaneously, a precision pulser would be counted by a scaler and would be used as a clock. Standard electronics and data acquisition system would be used. The γ-ray spectrum and the counts of the high precision pulser would be acquired for successive intervals of 15 minutes duration and then written on a computer disk. This would be followed by an automatic reset of the scalers, the erasure of the spectra from the spectrum buffer and the start of data collection for the next 15 minute interval. In this way the counting would continue for 8 hours at a stretch and then intermittently for another 20 hours. The live time of the counting system would increase with time as the ^{110}Sn source (half-life ~4.2 hours) would cool down. However the ratio of the peak areas of 280 keV γ-ray line produced due to the electron capture of ^{110}Sn nuclei and the sum of the peak areas of 1173 keV and 1332.5 keV γ-ray lines from ^{60}Co would be independent of the live time of the counting system and this ratio would be monitored with time. There should be no radioactive contaminant in ^{110}Sn beam and the γ-ray peaks of 280 keV (from ^{110}In), 1173 keV and 1332.5 keV (from ^{60}Co) should be free from any background peak. The intensity of any accompanying stable beam with the radioactive ^{110}Sn beam should be ≤ 10^3 ions/sec so that the lattice damage done by any such beam at the depth where ^{110}Sn would be implanted should be negligible (< 0.001 vacancies /ion/angstrom). Then the entire experiment would be repeated once starting with the implantation on Pd and Pb foil.

3. Experimental equipment:

We propose to use HIE-ISOLDE 2nd beam line (other than MINIBALL line) for implantation runs. The counting would be done later using a high efficiency HPGe or CLOVER detectors in a low background counting room. A standard ^{60}Co source (20-50 μC strength) is required. A precision pulser would be used for time keeping. Standard electronic and a data acquisition system will be required.

Summary of requested shifts:

The aim of the work is to achieve high-precision measurements of the half-life of ^{110}Sn implanted in large and small lattice spaces. It would provide information about the effect of compression on electron capture nuclear decay rate. The uncertainty in the measurement of the half-life would be kept within 0.05% to look for any change of decay rate under compression.

We request 5 shifts for this experiment. Each shift would be 8 hours long.

^{110}Sn can be obtained from HIE-ISOLDE with an intensity of >10^5 ions/sec and energy =2.2 MeV/A. The number of ^{110}Sn ions implanted after 8 hour run ≈2.8×10^9 ions. Since the energy of the ^{110}Sn beam is much below Coulomb barrier for the nuclear reactions with the nuclei of the catcher foils or beam line material (iron), the radioactive dose of the catcher foil would only come from the implanted ^{110}Sn ions and is expected to be ~3.5 μC. We need 1-1.5 hours to take out the implanted foil, put in another foil and start the next implantation run. The total number of 280 keV photons emitted initially from the source in 15 minutes ≈ 10^8. The total number of counts in the photo-peak of high efficiency HPGe detector in 15 minutes ≈5×10^6 counts at the beginning of the counting. The singles count rate in each γ-ray detector would be kept below 10000 counts per sec to enable good energy resolution of HPGe

detectors. The dead-time would be manageable as we shall take singles spectra. The effect of dead time of the data acquisition system would be cancelled by monitoring the ratio of the peak areas of 280 keV γ-ray line and ^{60}Co γ-ray lines with time.

The measurements would be done for 2 targets (Pd and Pb) and then repeated.

We shall need 4-5 hours of initial setup time.

We shall need 5 shifts for our implantation run.

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Appendix

Part of the Choose an item.	Availability	Design and manufacturing
HIE-ISOLDE 2nd beam line other than MINIBALL line) for implantation experiment. A low background room for counting will be required. High efficiency HPGe or CLOVER detectors or MINIBALL array along with standard electronics and data acquisition system will be required.	☒ Existing	☒ To be used without any modification
		DESCRIPTION OF THE PROPOSED EXPERIMENT The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*
[Part 1 of experiment/ equipment]	☐ Existing	☐ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[Part 2 experiment/ equipment]	☐ Existing	☐ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]		

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazards			
	[Part 1 of the experiment/equipment]	*[Part 2 of the experiment/equipment]*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	Standard ISOLDE vacuum		
Vacuum			
Temperature	Room temperature		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	None		
Electrical and electromagnetic			
Electricity	Standard requirement		
Static electricity	No		
Magnetic field	None		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			

Target material	Pb, Pd		
Beam particle type (e, p, ions, etc)	^{110}Sn		
Beam intensity	~10^5 particles/sec		
Beam energy	2.2 MeV/A		
Cooling liquids	None		
Gases	None		
Calibration sources:	☐		
• Open source	☐		
• Sealed source	☐ [ISO standard]		
• Isotope	^{60}Co sealed source		
• Activity	(20-50)μC		
Use of activated material:			
• Description	☐ ^{110}Sn implanted Pb and Pd foils		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope	^{110}Sn		
• Activity	<10μC		
Non-ionizing radiation			
Laser	No		
UV light	No		
Microwaves (300MHz-30 GHz)	No		
Radiofrequency (1-300MHz)	No		
Chemical			
Toxic	Pb foils		
Harmful	[chemical agent], [quantity]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	No		
Irritant	No		
Flammable	No		
Oxidizing	No		
Explosiveness	No		
Asphyxiant	No		
Dangerous for the environment	No		
Mechanical			
Physical impact or mechanical energy (moving parts)	No		
Mechanical properties (Sharp, rough, slippery)	[location]		
Vibration	[location]		
Vehicles and Means of Transport	None		
Noise	No		
Frequency	[frequency],[Hz]		
Intensity			
Physical			
Confined spaces	No		
High workplaces	No		
Access to high workplaces	No		
Obstructions in passageways	No		
Manual handling	[location]		
Poor ergonomics	[location]		

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*

1 kW