## EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time of Flight Committee

# Precision measurement of the half-life of <sup>109</sup>In in large and small lattice environments

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#### Abstract

We propose to undertake high precision measurements of the half-life of <sup>109</sup>In in large and small lattice environments to study the effect of compression on the electron capture nuclear decay rate. Such studies are of general interest having implications in many areas ranging from astrophysics to geophysics. 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 best available density functional calculations as obtained from TB-LMTO or WIEN2K codes. The proposed experiment should generate more data thus clarifying the experimental situation.

Requested shifts: 3 shifts

### 1 Motivation:

The study of the change of nuclear decay rate in different environments has fundamental significance and applications in many areas such as nuclear physics, condensed matter physics, astrophysics, geophysics etc. Among the different types of nuclear decays, 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 and so it is most susceptible to the surrounding environment, because the change of even the valence electron configuration should slightly modify the electron density at the nucleus. Since the surrounding environment can usually only change the configuration of the valence electrons, the effect of the environment should be most prominent on the electron capture rate of <sup>7</sup>Be (atomic configuration: 1s<sup>2</sup>2s<sup>2</sup>). The electron capture decay rate of <sup>7</sup>Be in different media has been extensively studied [1-7] and qualitatively understood in terms of the electron affinity of the surrounding medium. If the electron affinity of the surrounding medium is high, <sup>7</sup>Be loses more of its valence 2s electrons and so the electron density at the <sup>7</sup>Be nucleus drops decreasing the electron capture decay rate of <sup>7</sup>Be. Quantitative calculations [4,8] using density functional code TB-LMTO [9] also give reasonable agreement with the experimental results in many cases except when Be is implanted in lattices having very small open spaces.

On the other hand, the change of electron capture nuclear decay rate under compression is not at all well studied, although this is a very important topic. During the core collapse of the massive stars, the electron capture rate by the nuclei is expected to increase initially and the process plays an important role in the creation of heavy elements. The electron capture nuclear decay is also 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 [10] by the electron capture of <sup>40</sup>K in the core alone and this heat production is important in determining the thermal and tectonic evolution of the earth. 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. Regarding the experimental data, W. K. Hensley et al. [11] studied the change of electron capture decay rate of <sup>7</sup>BeO under compression and found that the decay rate increases by about 0.6% at a pressure of 270 kbar. Recently Liu and Huh [12] observed that the electron capture decay rate of <sup>7</sup>Be(OH)<sub>2</sub> gel increases by about 0.8% at a pressure of around 270 kbar. These results disagree completely with the standard density functional calculations such as TB-LMTO [9] and WIEN2K [13,14] codes which predict less than 0.1% increase of the electron density at the <sup>7</sup>Be nucleus due to the application of 270 kbar of pressure resulting in about 9% reduction of the volume of <sup>7</sup>BeO lattice. Both TB-LMTO and WIEN2K perform rigorous calculations with the valence electrons (2s orbital in the case of 'Be atom). However they apparently treat inner core orbital wave functions less rigorously. Although they consider 1s wave function of <sup>7</sup>Be in the lattice environment, the same boundary condition as that of a free atom, namely that the 1s wave function becomes zero at infinity, is applied. In the cases of the compression

of the larger many-electron radioactive (electron capturing) atoms such as <sup>40</sup>K, <sup>109</sup>In, <sup>110</sup>Sn etc., the increase of the electron density at the nucleus is generally expected to be very small, because the overlap of higher orbital valence electrons (such as 4s, 5s etc.) at the nucleus is exceedingly small. K. K. M. Lee and G. Steinle-Neumann performed [13,14] WIEN2K density functional calculations to calculate the increase of electron density at <sup>40</sup>K nucleus due to the compression of <sup>40</sup>K<sub>2</sub>O and found that the electron density at the <sup>40</sup>K nucleus should increase by < 0.01% even at a pressure of 500 kbar. However the increase of the electron density at the nucleus could be much higher, if the inner orbitals of the electron-capturing atom would also be compressed increasing the electron density at the nucleus. It is well known [15,16] that the eigenstate energies of the inner orbitals of an atom increase under spatial confinement. This compressional effect might also increase the electron density of the inner orbital electrons at the nucleus thus increasing the decay rate of an electron-capturing radioactive nucleus. At present, there is only one set of experimental data for 109 In and 110 Sn showing such an increase of the electron capture nuclear decay rate under compression. It was found [17] that the orbital electron capture rates of <sup>109</sup>In and <sup>110</sup>Sn increased substantially by (1.00±0.17)% and (0.48±0.25)% respectively when implanted in the smaller Au lattice compared to implantation in a larger Pb lattice. The modification of the valence (5s) electronic orbital of <sup>109</sup>In and <sup>110</sup>Sn should have negligible effect (10<sup>-5</sup>%) on the electron density at the <sup>109</sup>In or <sup>110</sup>Sn nucleus and actually the electron density at the nucleus should decrease very slightly when implanted in Au lattice because of the high electron affinity of Au. However because of their large sizes, both indium and tin atoms should experience significantly higher compression in the small Au lattice compared to that in the large Pb lattice. In fact, according to TB-LMTO density functional code, the difference in energy between a <sup>109</sup>In or <sup>110</sup>Sn ion implanted in an Au as compared to a Pb lattice should be much higher than what could be achieved for a <sup>7</sup>Be ion by applying an external pressure of 270 kbar. The effect of compression of the inner electronic orbitals of 109 In and 110 Sn ions in the smaller spatial confinement of Au lattice could increase the electron density at the nuclei and the corresponding electron capture rates. So the observed increase (~1%) of the electron capture decay rate of <sup>109</sup>In and <sup>110</sup>Sn in Au compared to when implanted in Pb is probably providing us information regarding the compression of the inner electronic orbitals when the ions are implanted in the smaller spatial confinement of an Au lattice. The compression of the inner orbital electrons of an implanted electron-capturing atom is a new idea and it would be very interesting if such effect can be studied by measuring the increase of the electron capture decay rate in the spatial confinement of a small lattice.

The interstitial spaces available in platinum and tantalum lattices are smaller than those available in gold lattice. So the eigenstate energies of <sup>109</sup>In implanted in platinum and tantalum lattices become much higher and the expected increase of the electron capture decay rate of <sup>109</sup>In implanted in platinum and tantalum should be higher than that of <sup>109</sup>In implanted in lead by more than 1%. The experiment of ref [17] was done by implanting <sup>109</sup>In and <sup>110</sup>Sn ions along with all other radioactive ions

produced in the heavy ion reaction <sup>20</sup>Ne+<sup>93</sup>Nb at E(<sup>20</sup>Ne)= 80 MeV. The availability of a pure <sup>109</sup>In beam from ISOLDE, CERN should enable us to perform a much cleaner measurement of this important effect.

## 2. Description of the experiment:

We propose to use a pure  $^{109}$ In beam (having intensity  $\sim 10^7$  ions/sec; energy= 60 keV) from ISOLDE and implant the ions in a Pt, Ta and Pb foil one by one. The implantation target would be kept at a negative potential of -10 to -20 kV, so that the <sup>109</sup>In ions would be incident on the target foils with energy of 70-80 keV and typically penetrate over 100 Angstrom in the target foil. The existing chamber which can be attached to the GLM beamline, used by the experiment IS500 to retard the beam particles could be modified easily to accelerate the beam particles and increase its energy by an additional 10-20 keV. The surface of the implantation targets would be polished to remove any oxide layer and then immediately put in high vacuum. The implantation on each target would be for about 2 hours (half-life of <sup>109</sup>In being ~4.2 hours). There would be no nuclear reaction between 109 In and the nuclei of the catcher foil, because the Coulomb barrier is much higher than the incident energy. After implanting <sup>109</sup>In ions in a foil for 2 hours, the implanted foil along with a standard <sup>60</sup>Co γ-ray source would be counted by placing it in MINIBALL array of ISOLDE. The count rate of each detector of the MINIBALL array would be kept around 10,000-15000 counts per sec (on the average) enabling good energy resolution (FWHM $\approx$  2 keV at 660 KeV energy). The time keeping would be done by using a precision pulser. The composite  $\gamma$ -ray spectrum from the  $^{109}$ In source (that would emit 203 keV  $\gamma$ -ray photons due to the electron capture of  $^{109}$ In nuclei) and  $^{60}$ Co source (that would emit 1173 keV and 1332.5 keV  $\gamma$ -ray photons) would be acquired along with the counts from a precision pulser. The dedicated electronics and data acquisition system of MINIBALL array would be used. The  $\gamma$ -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 10 hours at a stretch and then intermittently for another 30 hours. The livetime of the counting system would increase with time as the <sup>109</sup>In source (half-life ~4.2 hours) would cool down. However the ratio of the peak areas of 203 keV  $\gamma$ -line produced due to the electron capture of  $^{109}$ In nucleus and the sum of the peak areas of 1173 keV and 1332.5 keV  $\gamma$ -lines from  $^{60}$ Co should be independent of the livetime of the counting system and this could be verified by monitoring the ratio of the peak areas of 1173 keV to 1332.5 keV γ-ray lines from <sup>60</sup>Co with time. Regarding the purity of the incident <sup>109</sup>In beam, there should be no radioactive contaminant with the beam so that the  $\gamma$ -ray peaks of 203 keV (from <sup>109</sup>In), 1173 keV and 1332.5 keV(from <sup>60</sup>Co) should be free of any background peak and the uncertainty of the area determination of the peaks should be only statistical. The intensity of <sup>109</sup>In beam should be around 10<sup>7</sup> ions per sec and it can be obtained by using the target material UC<sub>x</sub> with 1 µA of primary proton beam. There should not be any radioactive contaminant present in the beam. Regarding the presence of the stable contaminant in the  $^{109}$ In beam, the intensity of any accompanying stable beam should be less than  $10^5$  ions/sec so that the lattice damage done by any such beam at the depth where  $^{109}$ In would be implanted should be negligible (< 0.001 vacancies /ion/angstrom).

The measurements would be done by placing <sup>109</sup>In implanted platinum foil, tantalum foil and lead foil in MINIBALL array one by one after the respective implantation run. Then the entire experiment would be repeated once starting with the implantation on platinum, tantalum and lead foils.

## 3. Experimental equipment:

We propose to use GLM chamber used by the experiment IS500 and modify it for accelerating beam particles. MINIBALL array and its dedicated electronics and data acquisition system would be used to take singles data. A precision pulser would be used for time keeping.

## **Summary of requested shifts:**

The aim of the work is to achieve a high-precision measurement of the half-life of  $^{109}$ In in large and small lattice environments. The uncertainty in the measurement of the half-life would be kept within 0.15% to clearly see the effect.

We request 3 shifts for this experiment.

 $^{109}$ In can be produced by using a UC target and surface ionisation. ISOLDE SC yields give  $1.7\times~10^7$  ions/µc of protons and we assume this proton current for our proposal. The number of  $^{109}$ In ions implanted after a 2 hour run  $\approx 6\times 10^{10}$  ions. We need one and half hour to take out the implanted foil, put in another foil and start the next implantation run. The total number of 203 keV photons emitted from the source in 15 minutes  $\approx 10^9$ . The total number of counts in the photo-peaks of MINIBALL array in 15 minutes  $\approx 10^8$  counts at the beginning. The singles count rate in each HPGe detector of MINIBALL array will be  $\sim 10000-15000$  counts per sec on the average, although it would be higher at the beginning. The dead-time would be manageable as we shall take singles data.

The measurements would be done for 3 targets (Pt, Ta and Pb) and then all the measurements would be repeated.

We shall need 3 hours of setup time initially.

So we need total of 24 hours of beam time = 3 shifts.

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## Appendix

### **DESCRIPTION OF THE PROPOSED EXPERIMENT**

The experimental setup comprises: (name the fixed-ISOLDE installations, as well as flexible elements of the experiment)

Part of the Choose an item.	Availability	Design and manufacturing
A low background room for	Existing	To be used without any modification
counting. MINIBALL array , its		
dedicated electronics and data		
acquisition system will be used.		
GLM chamber will be modified to		
accelerate beam particles by an		
additional 10-20 keV.		
[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				
Vacuum	Standard ISOLDE vacuum			
Temperature				
Heat transfer				
Thermal properties of materials				
Cryogenic fluid	None			
Electrical and electromagnetic				
Electricity	-10 to -20 kV kV target			
	station.			
Static electricity		_	_	
Magnetic field	None	_		

Batteries			
Capacitors			
Ionizing radiation			
Target material	[material]		
Beam particle type (e, p, ions,	<sup>109</sup> In		
etc)			
Beam intensity	10 <sup>7</sup> ions/sec		
Beam energy	60 keV		
Cooling liquids	None		
Gases	None		
Calibration sources:			
Open source			
Sealed source	[ISO standard]		
Isotope	<sup>60</sup> Co, <sup>109</sup> In		
Activity	< 100 μ curie (produced		
Activity	initially; dropping rapidly)		
Use of activated material:	initially, dropping rapidly)		
Description			
	[dose][mSV]		
Dose rate on contact     and in 10 cm distance	[aose][msv]		
Isotope			
Activity			
·			
Non-ionizing radiation	T	1	
Laser			
UV light			
Microwaves (300MHz-30			
GHz)			
Radiofrequency (1-300MHz)			
Chemical		1	
Toxic	Pb foil		
Harmful	Pb foil		
CMR (carcinogens, mutagens			
and substances toxic to			
reproduction)			
Corrosive	None		
Irritant	None		
Flammable	None		
Oxidizing	None		
Explosiveness	None		
Asphyxiant	None		
Dangerous for the	None		
environment	l .		
Mechanical	L		
Physical impact or	None		
mechanical energy (moving			
parts)	News		
Mechanical properties (Sharp, rough, slippery)	None		
(Sharp, rough, slippery) Vibration	None		
Vehicles and Means of	None		
Transport	[location]		
	l	1	
Noise	Ι.,		
Frequency	No noise		
Intensity			
Physical			
Confined spaces	A low background counting		
	room		

High workplaces	None	
Access to high workplaces	Not required	
Obstructions in passageways	No	
Manual handling	[location]	
Poor ergonomics	[location]	

## 3.1 Hazard identification

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)

Negligible