# Precision measurement of the change of the half-life of $^{110}$Sn by confinement in the large and small lattices

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# BACKGROUND

Radioactive decay generally known to be independent of external environment, such as pressure.

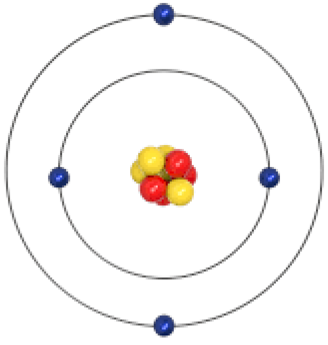
However, $e^-$ capture nuclear decay is susceptible to environmental change.

Electron capture nuclear decay rate

$$\lambda_{EC} = \frac{(E_\nu)^2}{\pi c^3 \hbar^4} g^2 \left|H_{fi}\right|^2 |\psi_e(0)|^2$$

$$|\psi_e(0)|^2 = |\psi_{1s}(0)|^2 + |\psi_{2s}(0)|^2 + \dots\ |\psi_V(0)|^2$$

External environment can affect $|\psi_V(0)|^2$



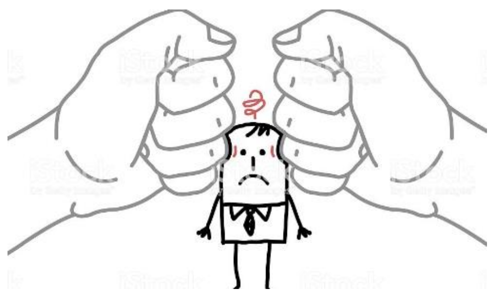
Effect of compression increases electron capture nuclear decay rate.
Measured increase of EC decay rate under compression
>> Theoretical expectations



Effect of compression studied experimentally by two different methods– Diamond Anvil Cell and Implantation in a small lattice

# Compression of $^7$Be atom by confining in a small lattice

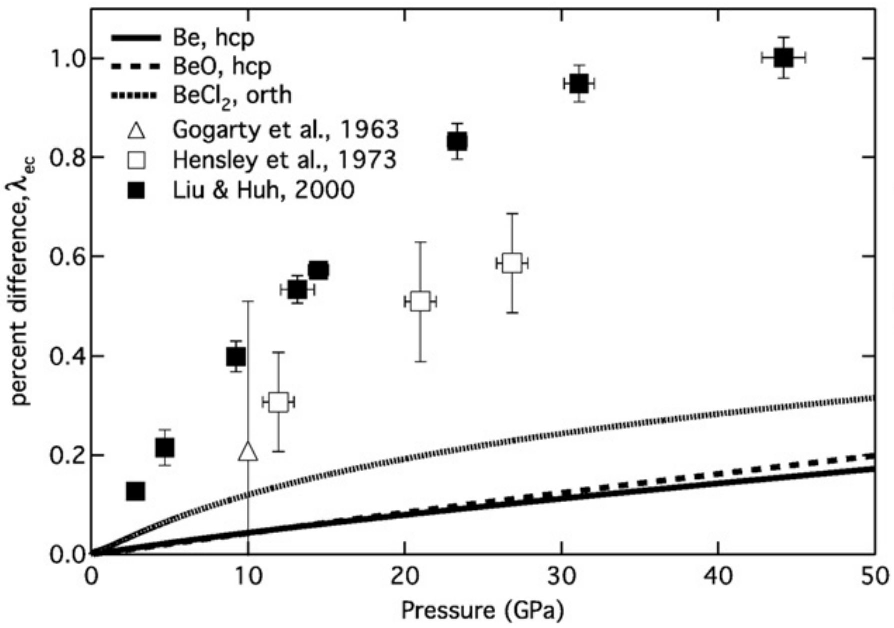
$^7$Be ion compressed when implanted in a small lattice such as Palladium (Pd).
Lattice constant = 3.9Å
Compared to
when implanted in a large lattice such as lead (Pb).
Lattice constant = 5Å
Both Pd and Pb have low and similar electron affinity.
Observed increase of decay rate of $^7$Be in Pd could be attributed to compression.

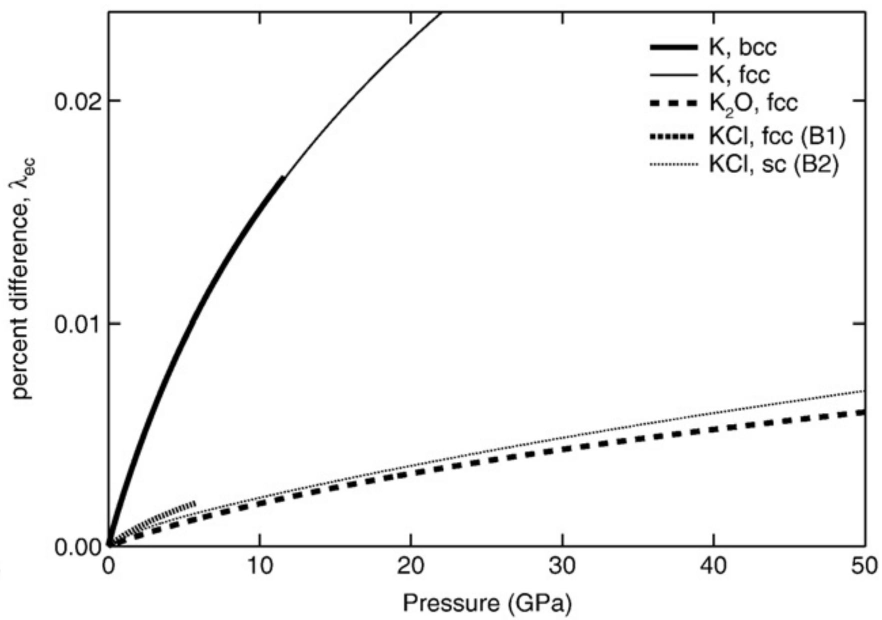


Measured increase of decay rate of $^7$Be in Pd w.r.t Pb is $\Delta\lambda/\lambda=(0.82\pm0.16)\%$
[A. Ray *et al.*, Phys. Rev C **101**, 035801 (2020)]
WIEN2K predicts ~0.2% increase of decay rate of $^7$Be in Pd.
Large discrepancy between measurements and DFT calculations

# Effect of Compression on EC decay

Calculations by Lee and Steinle-Neumann, EPSL 267, **628** (2008)




We performed WIEN2k density functional calculations for Sn in small Pd lattice (lattice constant = 3.89Å) versus large Pb (lattice constant = 4.95Å).

No observable change expected. Change ~$10^{-3}$ %. Inclusion of finite nucleus effect could increase the effect by a factor of 4, but still very small.
Earlier measurement found decay rate of $^{110}$Sn in Au (lattice constant=4.08Å) faster than in Pb (lattice constant=4.95Å) by (0.48±0.25)%. For $^{109}$In, increase = (1.0±0.2)%.
[A. Ray *et al.*, Phys. Lett. B **679**, 106 (2009)]

# Other theoretical possibilities

Observation of anomalous increase of radioactive decay rate under compression not limited to EC decay only.
$\alpha$-decay rate of $^{221}$Fr increases by (0.42±0.21)% in W lattice (lattice constant=3.16Å) versus Si lattice (lattice constant=5.43Å). Theoretical expectation ~ $(10^{-3} - 10^{-4})$%.
[H. B. Jeppesen et al., Eur. Phys. J. A **32**, 31 (2007). (CERN ISOLDE experiment)]

**Quantum Anti-Zeno effect:**
Decay nonexponential at initial time.
Nonexponential decay time for $^{110}$Sn decay $t_{ne} \sim \hbar/E_{decay} \approx 10^{-21}$ s for $E_{decay}$ = 0.631 MeV.
Increase of kinetic energy of electrons of Sn atom in Pd versus Pb is $\Delta E \approx 350$ eV.
Increase of decay rate due to Quantum Anti-Zeno effect $\sim(\Delta E/ E_{decay})$=0.05%.
[Kofman and Kurizki, Nature **405**, 546 (2000).]

Initial nonexponential decay time $t_{ne}$ could be much longer $\geq 10^{-21}$ s in certain models of quantum measurement that considers recording by the environment. [Fonda et al., Rep. Prog. Phys. **41**, 587 (1978); Zurek, Rev. Mod. Phys. **75**, 715 (2003); Ray and Sikdar, Phys. Rev C **94**, 055503 (2016).]

Effect of long $t_{ne}$ on quantum anti-Zeno effect not yet probed.

# Physics Motivation for measuring change of EC decay rate under compression with a high precision

Electron capture nuclear decay rate plays an important role in the creation of heavy elements during the merger of neutron stars and core collapse of supernova.
Significant heat produced by the electron capture of $^{40}K$ at the earth's core.
Very high compression involved in these EC decays.
We rely on calculations for determining EC decay rates in these extreme environments.

Anomalously large increase of EC decay rate under compression reported in lab expt.

*Reported large increase of EC decay rate under compression (contrary to the theoretical expectations) and the importance of EC decay in astrophysical and geophysical scenarios provide the* ***Physics Motivation*** *for measuring change of EC decay rate under compression with a high precision. Establish the effect of compression on EC decay conclusively.*

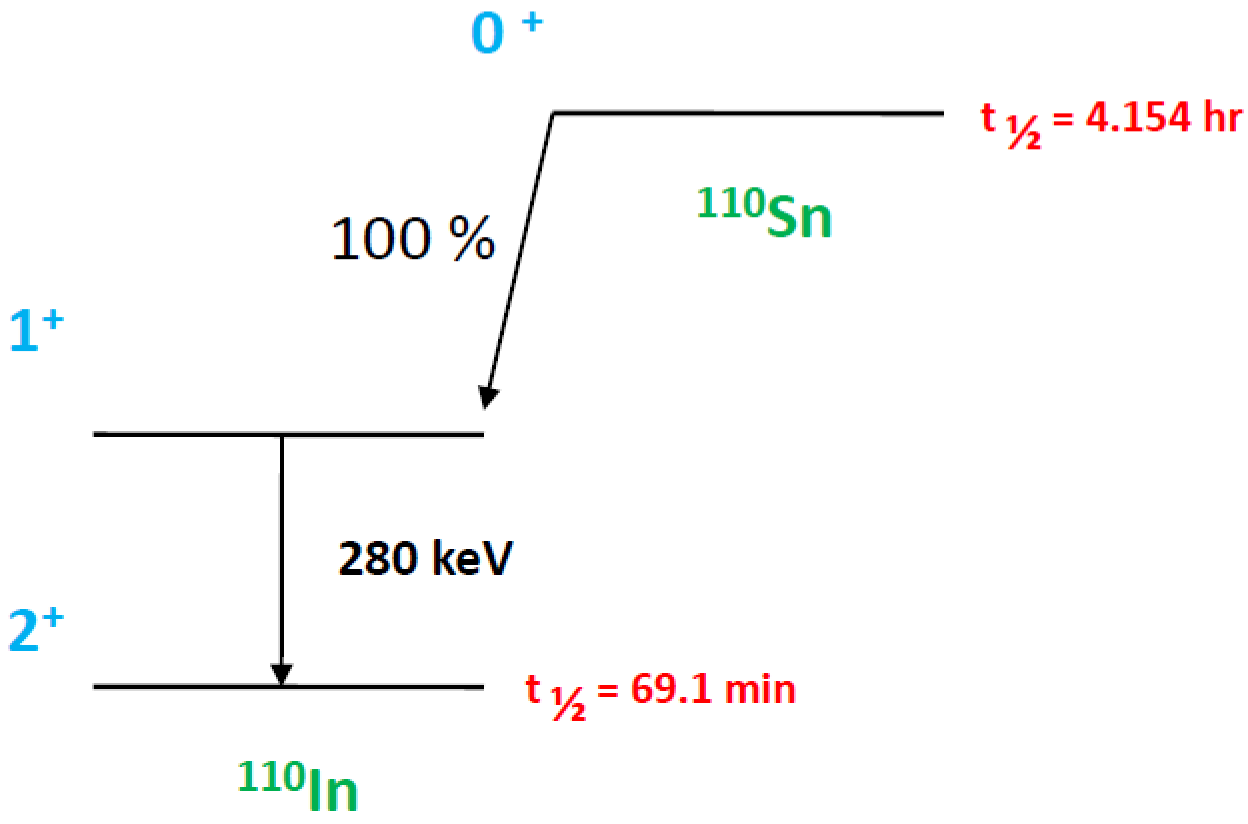
$^{110}Sn$ easy beam at CERN ISOLDE. Half-life= 4.15 hours (decay scheme shown).
Easy to monitor decay over many (say 10) half-lives.

To study the effect of compression, change of decay rate of $^{110}Sn$ implanted in a small Pd lattice and large Pb lattice would be measured with *a high precision of 0.05%*.



**GOAL:** Establish the reported increase (0.48±0.25)% of decay rate by a high statistical confidence level or show that such an anomalous increase of EC decay rate does not exist.

# Decay Scheme of $^{110}Sn$



# Proposed Experiment

$^{110}$Sn ions (lifetime ≈4.15 hours) with intensity ≈ $10^7$ ions/s and energy = 2.2 MeV/A would be implanted in Pd and Pb catcher foils (25 μm thick) one by one.
Implantation at a depth of 10-15 μm.
No surface effect.
Electron affinity of Pd is = 0.56 eV and Pb is = 0.37 eV.
Energy of $^{110}$Sn beam below Coulomb barrier for nuclear reactions with Pb or Pd.

Implantation on each catcher foil for >2 hours.
Implanted Pb and Pd foil along with a $^{60}$Co γ-ray source to be placed one by one before a high efficiency HPGe detector in a low background room.

Count for 15 minutes, then save spectrum, reset and start with the other implanted foil.
Count for the next 15 minutes, interchange foils and continue in this way for 41.5 hours.

Complete measurements for Pb and Pd foils. Repeat measurements once.
Time keeping by a calibrated pulser.
Monitor ratio of 280 keV γ-ray line to 1332.5 keV γ-ray line from $^{60}$Co with time to cancel out dead time and pile-up effects.
Measure Relative change of decay rate of $^{110}$Sn in Pb and Pd foils.

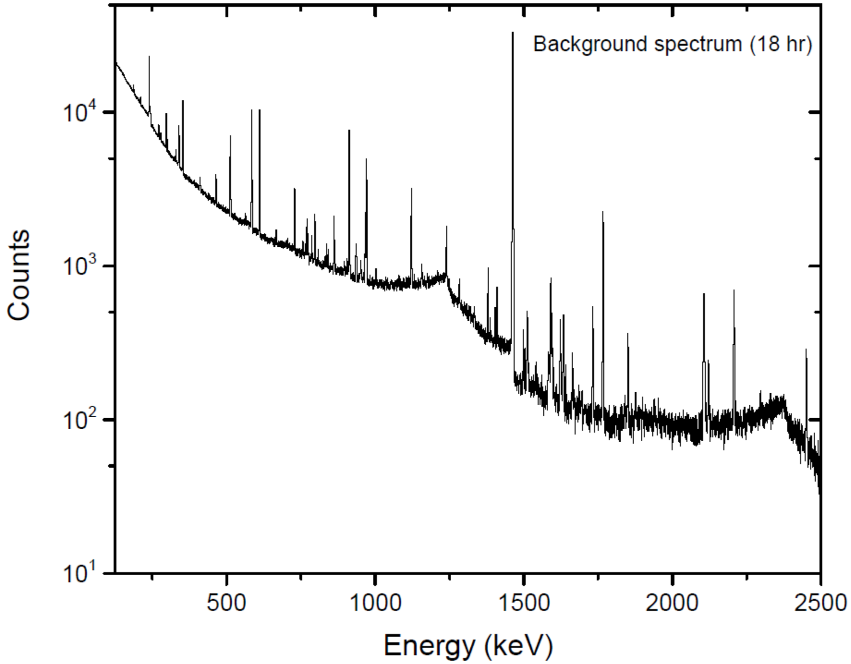
# Estimation of experimental precision

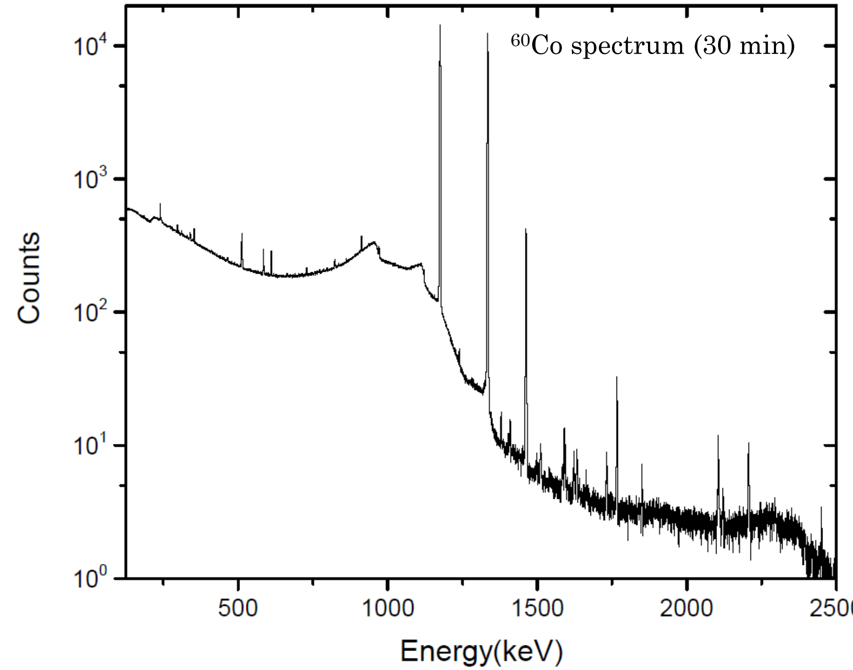
2 hours of implantation in Pd or Pb foils (25 micron thick) with a pure $^{110}$Sn beam of intensity ~$10^7$ ions/s and energy = 2.2 MeV/A.
Total number of implanted ions =$6\times10^{10}$ $^{110}$Sn ions.

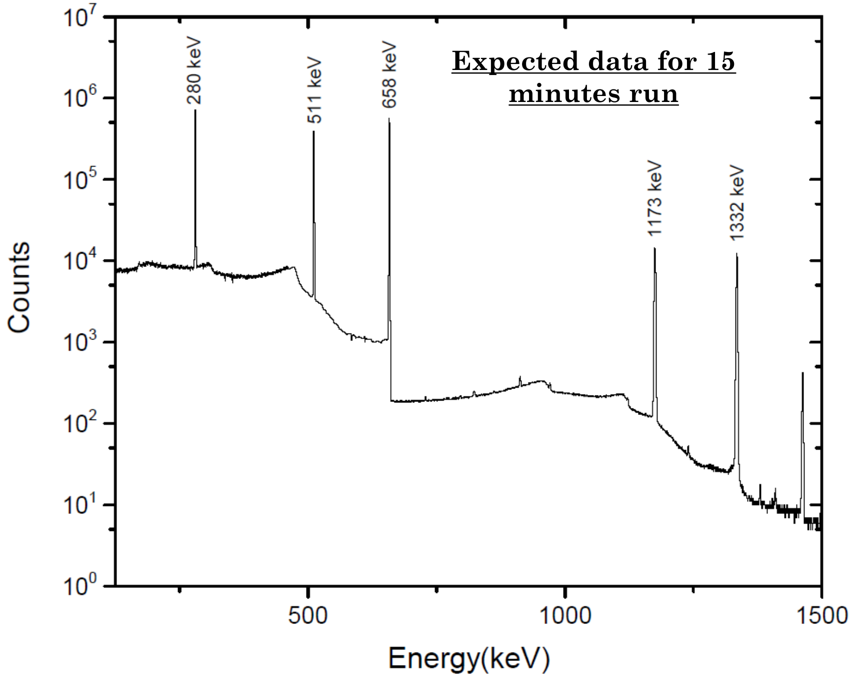
Counting would start 4 hours after the completion of implantation.
Number of $^{110}$Sn ions =$3\times10^{10}$
Number of decays in 15 minutes is = $8.3\times10^{8}$
Number of counts in 280 keV γ-ray photo-peak in 15 minutes is ≈$3\times10^{6}$ at the beginning of counting.

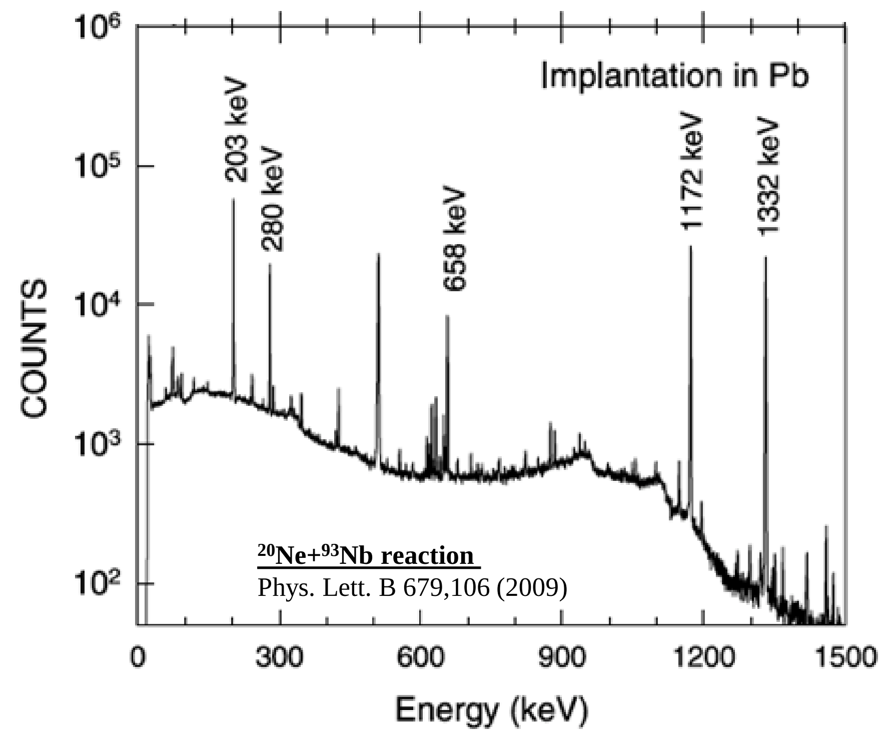
A $^{60}$Co source counted with it.
Total counts in the photo-peaks of 1173 keV and 1332.5 keV γ-ray lines in 15 minutes would be made ≈$2\times10^{5}$

**Simulation of γ-ray spectrum**
Background γ-ray spectrum taken for 18 hours in a low background room using a high efficiency (80% efficiency) HPGe detector covered with the lead bricks.
γ-ray spectrum of a $^{60}$Co source taken with the same high efficiency detector.
Compton background of 1173 keV and 1335.5 keV γ-ray lines determined.
280 keV γ-ray Gaussian peak simulated and put in.
658 keV and 511 keV γ-rays would be produced from the decay of isomeric $^{110}$In.
Compton backgrounds of these peaks determined from suitable source spectra.


Background spectrum (18 hr)
Counts
Energy (keV)

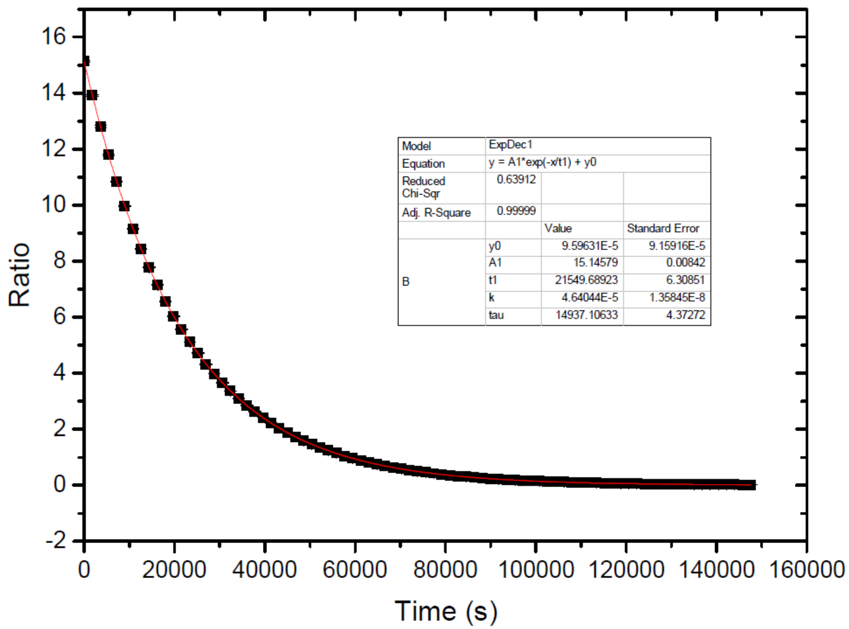

$^{60}$Co spectrum (30 min)
Counts
Energy(keV)

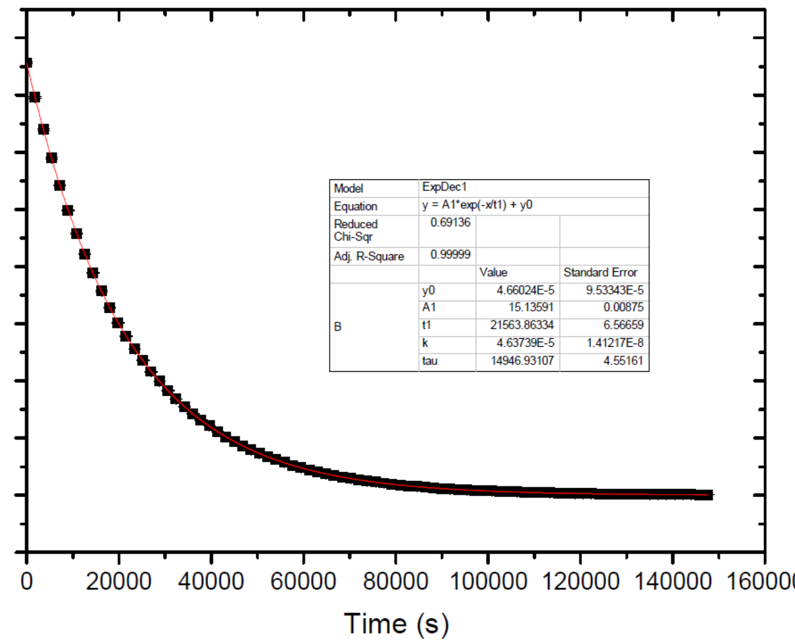

Expected data for 15 minutes run
280 keV
511 keV
658 keV
1173 keV
1332 keV
Counts
Energy(keV)


Implantation in Pb
203 keV
280 keV
658 keV
1172 keV
1332 keV
COUNTS
Energy (keV)
$^{20}$Ne+$^{93}$Nb reaction
Phys. Lett. B 679,106 (2009)

# Simulation of exponential decays by Monte Carlo technique

Ratio of 280 keV γ-ray peak from $^{110}$Sn to the sum of 1173 keV and 1335.5 keV γ-ray peaks from $^{60}$Co monitored over 10 half-lives of $^{110}$Sn. (Monte Carlo simulations done)
Only statistical error considered, because ratio of peak to valley ~100.
Half-life of $^{110}$Sn taken as 4.15 hours and 0.05% longer in the other cases.
Initial count rate ≈20,000 counts/sec



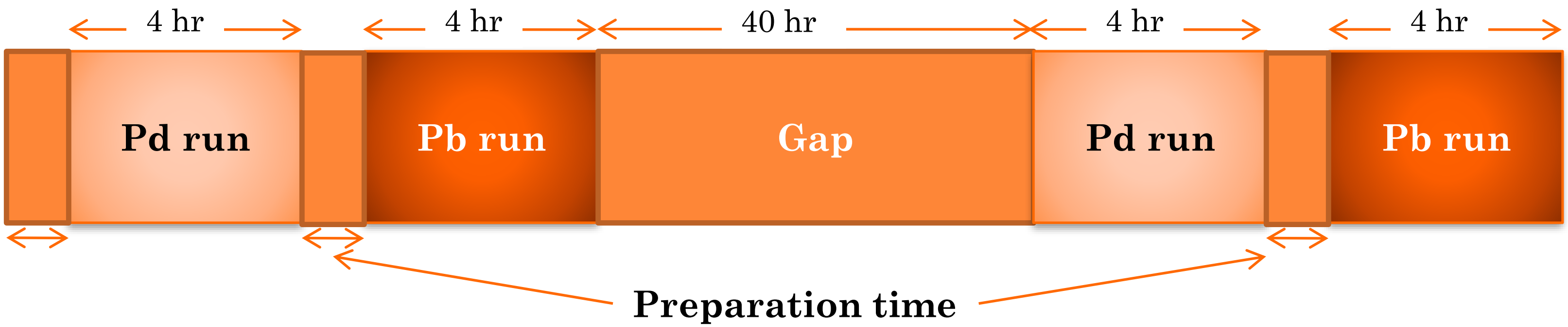




Difference of half-life from the two exponential data sets: (half-life≈14946.93 sec) =(9.83±6.31) sec
Percentage change of half-life between two data sets is= (0.065±0.042)%

**Precision of measurement =0.04%.**

# Requested Shifts

Assuming $^{110}$Sn beam current ~$5\times10^6$ ions/s
Implantation time on one target= 4 hours.




Total implantation time on 4 targets (considering repeat measurements) is =4×4=16 hours

Assuming time to change target = 2 hours
Total target changing time = 2×3=6 hours.
Total time= (16+6) hours = 22 hours.
**Requesting 3 shifts ( each shift 8 hours long)**

After 1st shift, a gap of 40 hours should be given to complete counting and start repeat implantation.

**4-5 hours time required for initial setup.**


thanks

# EXTRA SLIDES FOR DISCUSSIONS

# Requirement of Beam Purity

$^{110}$Sn beam would contain $^{110}$In impurity.
$^{110}$In→$^{110}$Cd emits 657.8 keV γ-ray. Compton edge at 280 keV.
Contamination could be reduced by electronic gating.

**Want radioactive contaminant $^{110}$In to be ≤1% of $^{110}$Sn beam intensity.**

Stable beam contaminants could produce additional lattice damage.
Might produce nuclear reaction.

Heavier (stable) contaminants could be more acceptable.

Prefer **$^{110}$Sn ($27^{+}$)** beam with Xe, Ce, La stable contaminants.

**Want heavy stable contaminants ≤1% of $^{110}$Sn beam intensity.**

# Energy Resolution and Dead Time

Initial count rate ≈ 20,000 counts/sec

HPGe energy resolution not affected significantly.

Consider a VME (Versa Module Euro card) data acquisition system

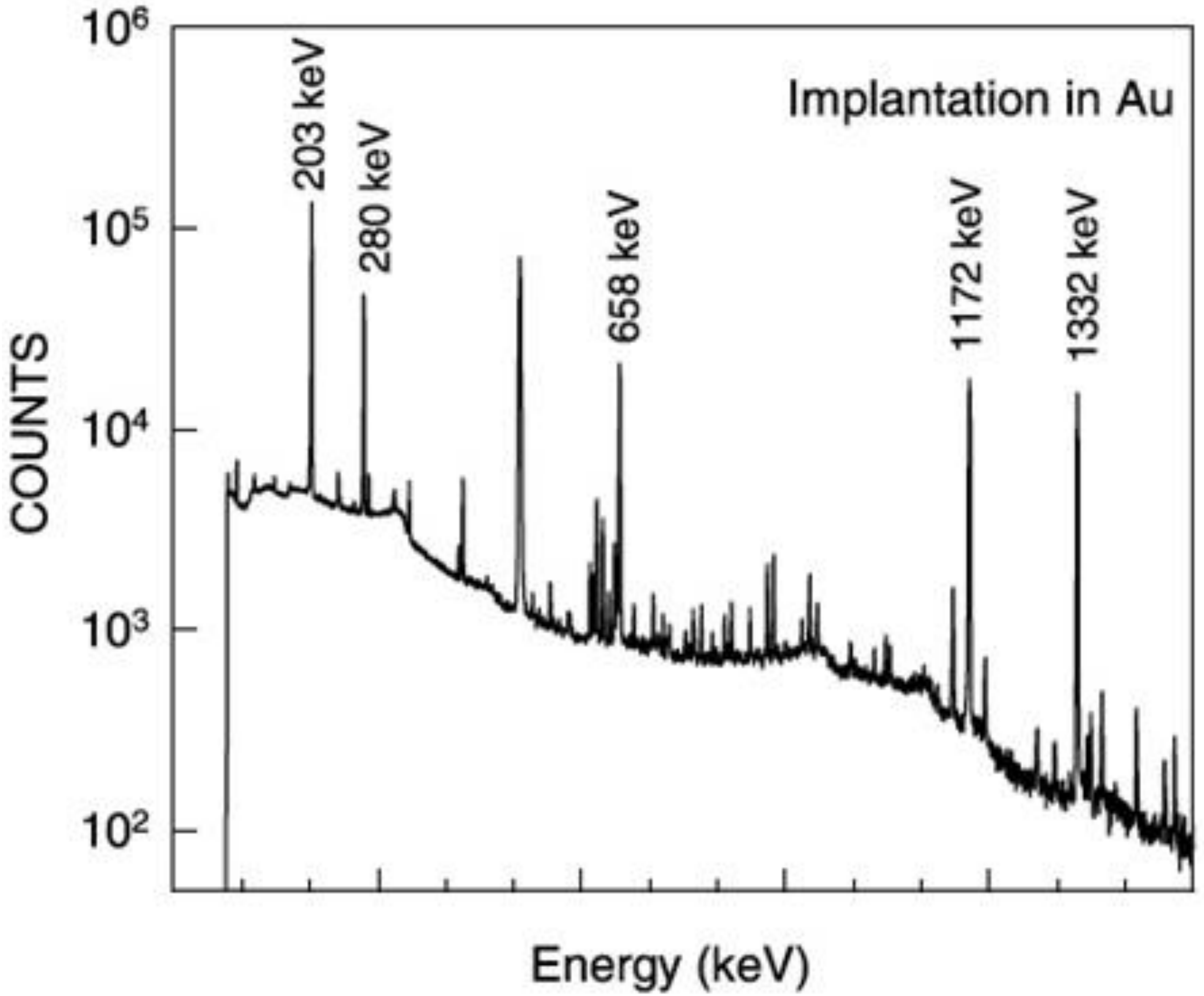
20,000 counts/sec means data rate = 0.24 MB/sec
Data rate that VME can handle = 5 MB/sec
VME ADC conversion time = 8 μs.

Let R be count rate and $\tau$adc conversion time.
Then Dead time $\tau_D = \frac{R\tau}{1+R\tau} \times 100\%$

So, $\tau_D = \frac{2\times10^4\times8\times10^{-6}}{1+2\times10^4\times8\times10^{-6}} \times 100\% = 13.8\%$

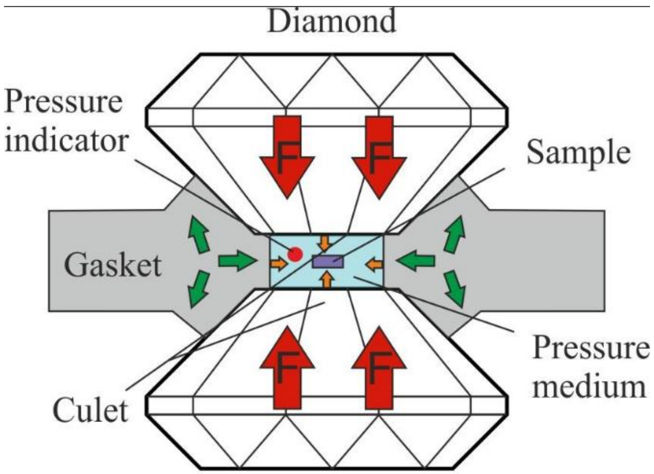
$\tau_D$ decreases with time as count rate R drops.

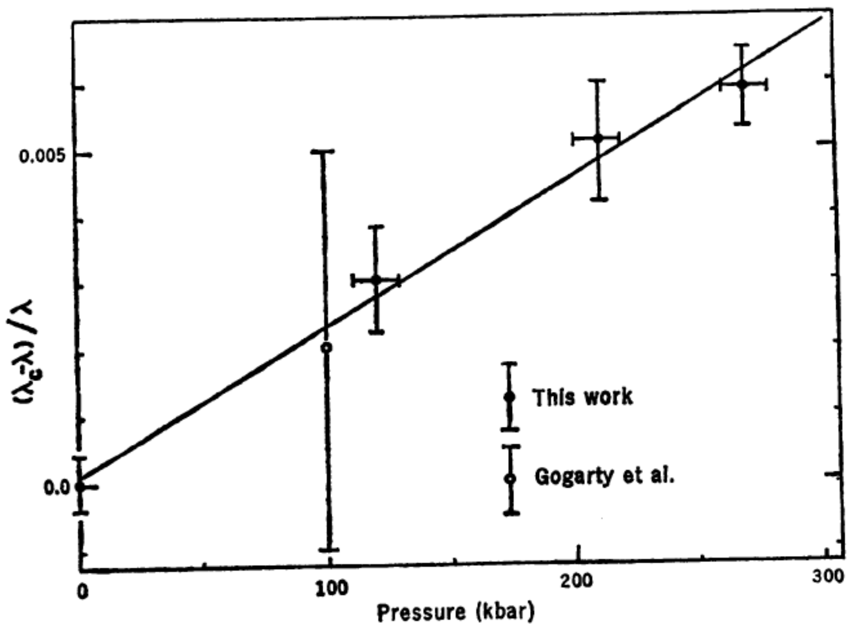
**Monitor ratio of 280 keV peak area to $^{60}$Co peaks areas with time.**
**No dead time effect in the extracted half-life.**




$^{20}$Ne+$^{93}$Nb reaction, A. Ray et al., Phys. Lett. B**679**, 106 (2009).

# Compression of $^7$Be atom by applying external pressure on $^7$BeO lattice




W. K. Hensley *et al.*, Science 181, 1164 (1973)

$\frac{\Delta\lambda}{\lambda} = (2.2 \pm 0.1) \times 10^{-4} P$ , where P is the applied pressure in GPa.

Calculations underpredict experimental results by a factor of 3-5.
EPSL **267**, 628 (2008); Phys. Rev C **88**, 034608 (2013);
Phys. Rev C **90**, 019801 (2014).

# Reasons for performing $^{110}$Sn experiment at CERN HIE-ISOLDE

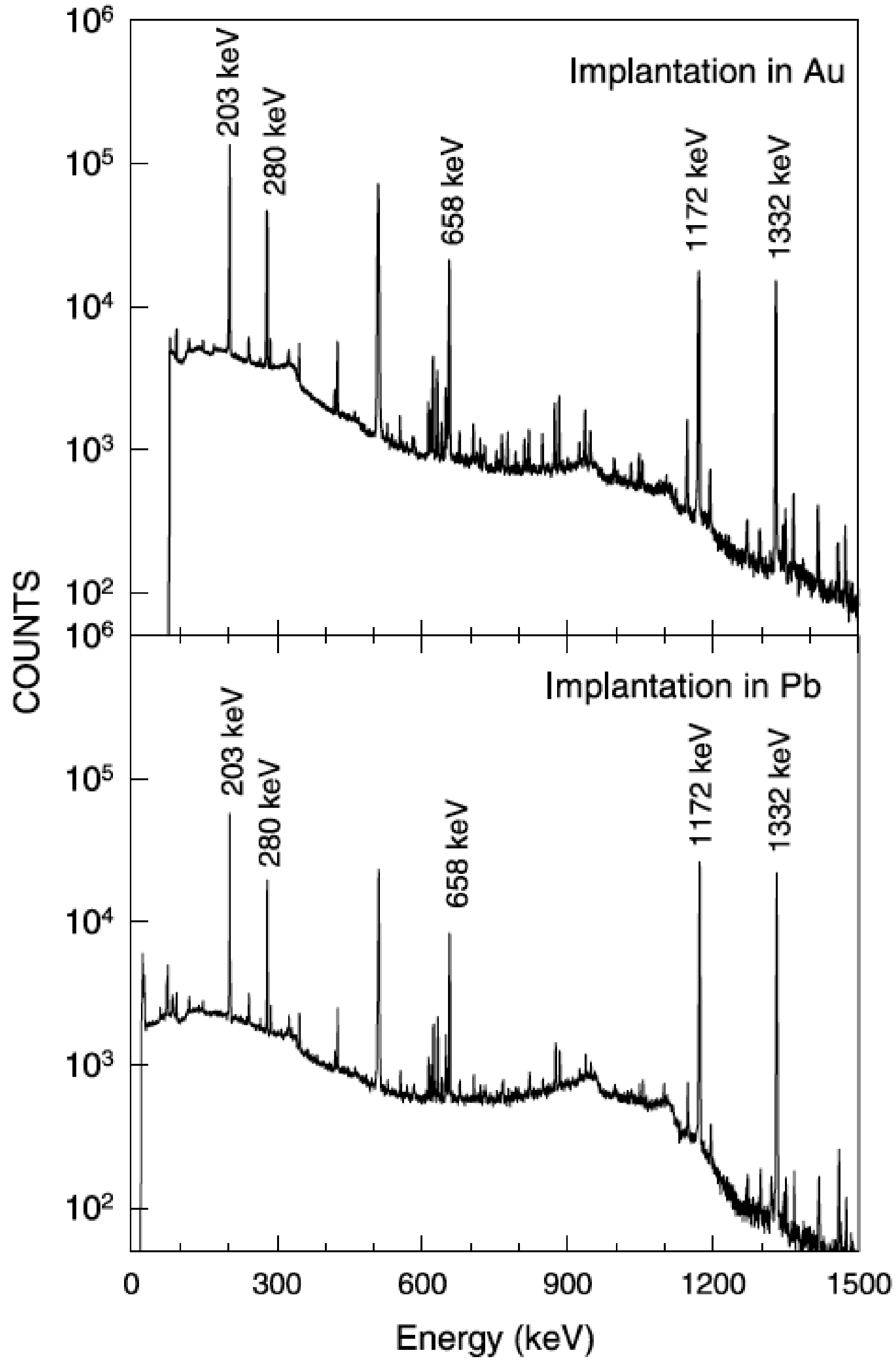
In the earlier experiment [$^{20}$Ne+$^{93}$Nb reaction at E($^{20}$Ne)=80 MeV],
ALL radioactive products implanted in Pb and Au catcher foils.

**Uncertainties in 280 keV$\gamma$-ray peak area determination**
Ratio of peak to valley for 280 keV $\gamma$-ray line was 12 in earlier expt.
Uncertainties in drawing background line.
Leads to $\approx$0.2% uncertainty in half-life determination.

**For higher precision experiment, plan to use**
Intense (~$10^7$ ions/s) energetic (2.2 MeV/A) $^{110}$Sn beam from HIE-ISOLDE.
No nuclear reaction. Most intense $\gamma$-ray peak expected at 280 keV.
Peak to valley ratio for 280 keV$\gamma$-ray line expected to be ~100.
Half-life determination with $\approx$0.05% uncertainty possible.

**Implantation in bulk region required**
50-60 keV$^{110}$Sn ions from CERN ISOLDE implanted at depths of (10±6) nm.
Surface effects.

$^{20}Ne+^{93}Nb$ reaction



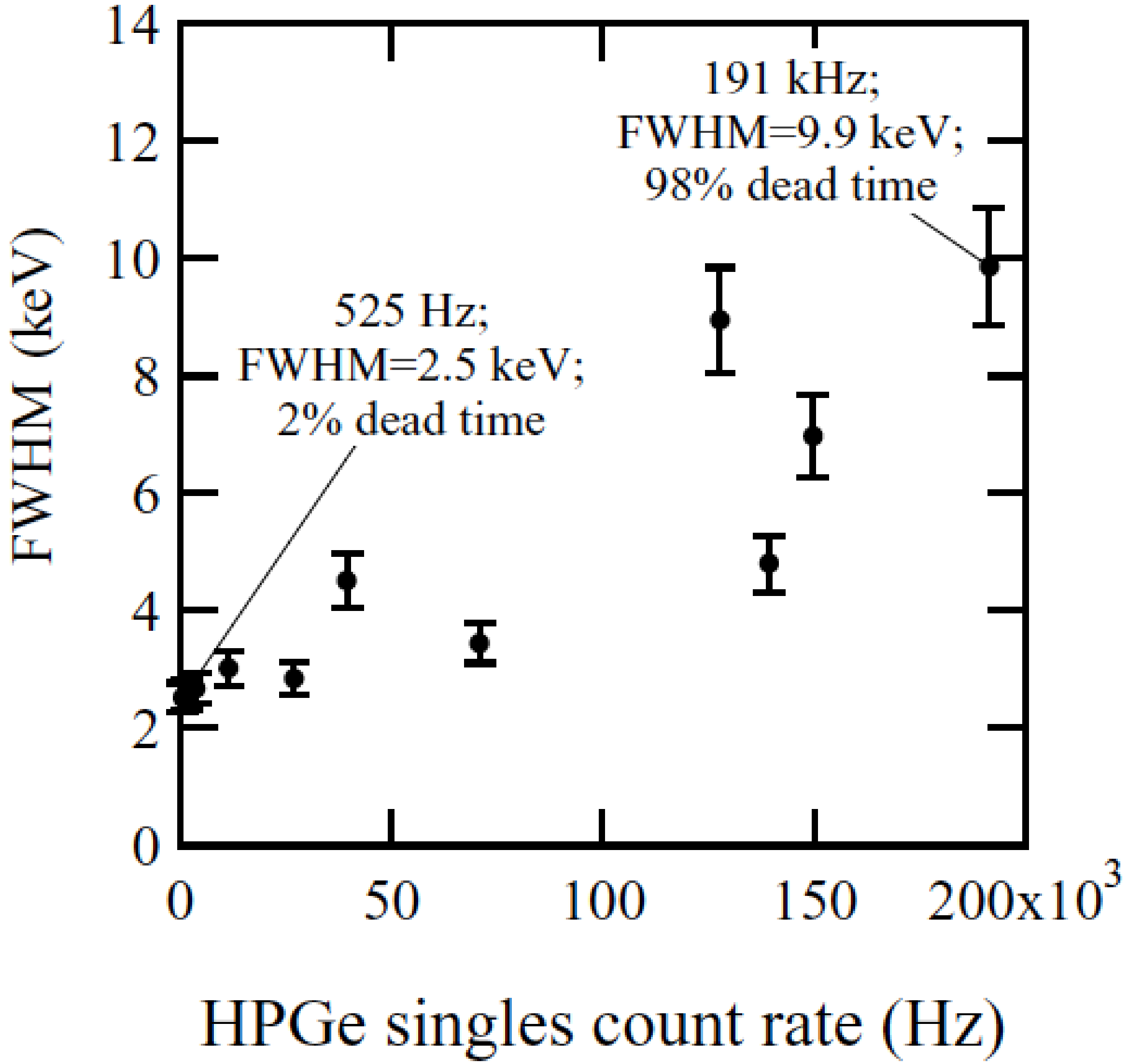
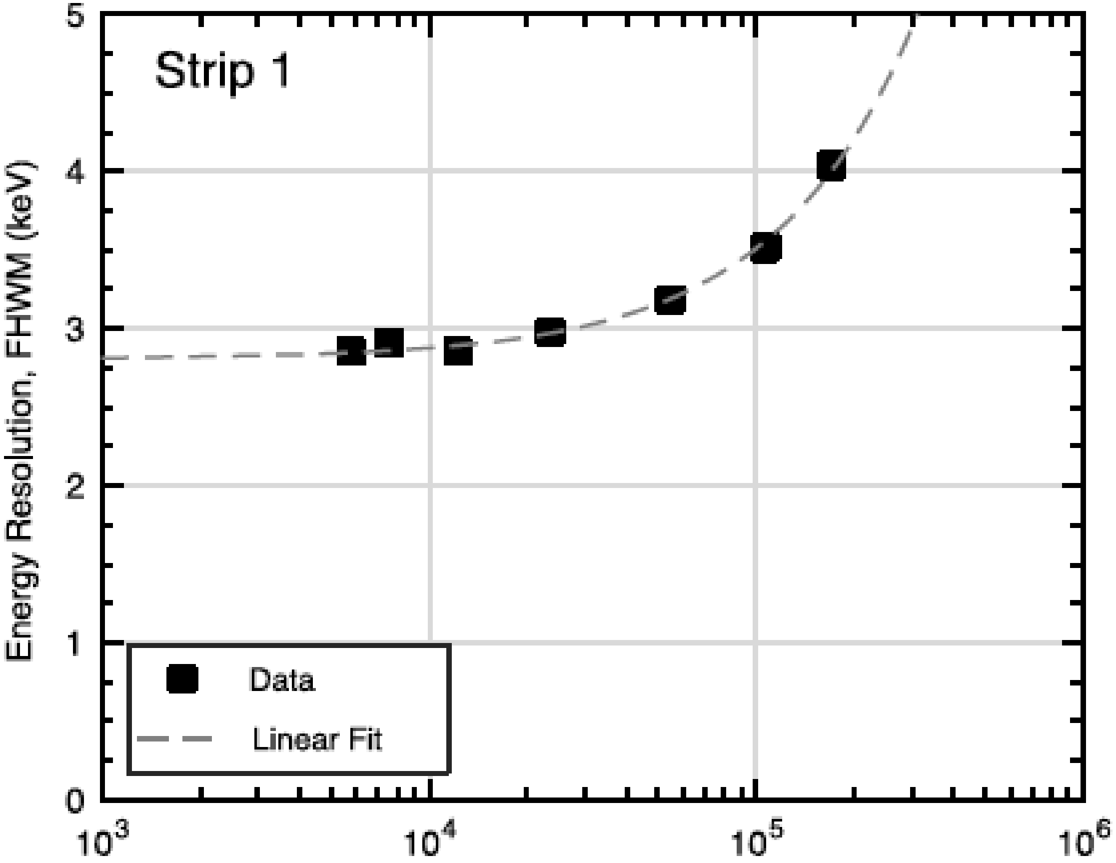



Figure 6. Energy resolution of 1173 keV Co-60 line in HPGe detector as a function of count rate.

J. C. Cooper and D. Koltick, IEEE




R. J. Cooper et al., Nucl. Inst. And Methods in Physics Research A **886**, 1 (2018)

Bibikov et al. [A. V. Bibikov *et al*., Phys. Rev. C **88** (2013) 034608] performed Hartree-Fock calculations for compression of $^{7}$BeO lattice. Underpredicted increase of $^{7}$Be decay rate under compression by a large factor.

Proposed structural phase transition of $^{7}$BeO lattice under compression to rock-salt structure to explain experimental observations.

X-ray study by Mori et al. [Y. Mori *et al*., Phys. Status Solidi B **241**, 3198 (2004)] did not show any structural phase transition below 100 GPa.

Ray and Das [A. Ray and P. Das, Phys. Rev. C **90** (2014) 019801] showed that structural phase transition to rock-salt structure would decrease (instead of increasing) $^{7}$Be decay rate.

Both DFT and Hartree-Fock calculations underpredict the experimental results by a large factor.

# Monte Carlo Techniques

## Exponential decay

Let τ be the lifetime of exponential decay. Then random decay time

$$t = -\tau \ln(u)$$

Where $u$ is a random number between 0 and 1.

## Gaussian Distribution

Let $u_1$ and $u_2$ be random numbers between 0 and 1.

$$v_1 = 2u_1 - 1$$
$$v_2 = 2u_2 - 1$$
$$r^2 = v_1^2 + v_2^2$$

If $r^2 > 1$, start over

if $r^2 < 1$, then

$$z_1 = v_1\sqrt{\frac{-2lnr^2}{r^2}}; \; z_2 = v_2\sqrt{\frac{-2lnr^2}{r^2}}$$



$z_i' = \mu + \sigma z_i$ gives a Gaussian distribution with a mean value $\mu$ and variance $\sigma^2$.

**Final stage evolution of massive star leading to supernova.**

8-10 solar mass star
After carbon burning, O-Ne-Mg core formed.
EC decay by $^{20}$Ne important.

EC decays of $^{54,56}$Fe, Ge play important role for core-collapse supernova.

$$\lambda_{e_{cap}}(T) = \text{const} \times \int_{E_{th}}^{\infty} \sigma(E_e, T) E_e p_e c f(E_e)$$

$$\sigma(E_e, T) = \sum_i \frac{(2J_i + 1)e^{-Ei/KT}}{G(Z, A, T)} \sum_f \sigma_{f,i}(E_e)$$

$$G(Z, A, T) = \sum_i (2J_i + 1)e^{-E_i/KT}$$

$$f(E_e) = \frac{1}{1 + exp\left(\frac{E_e - \mu_e}{KT}\right)}$$

$\mu_e$ depends on baryon density $\rho$ and proton fraction $Y_e$.

How to calculate chemical potential $\mu_e$

$$\rho Y_e = \frac{1}{\pi^2 N_A} \int_0^\infty \left[f_e\left(E_e - f_{e+}(E_e)\right)\right](p_e c)^2 d(p_e c)$$

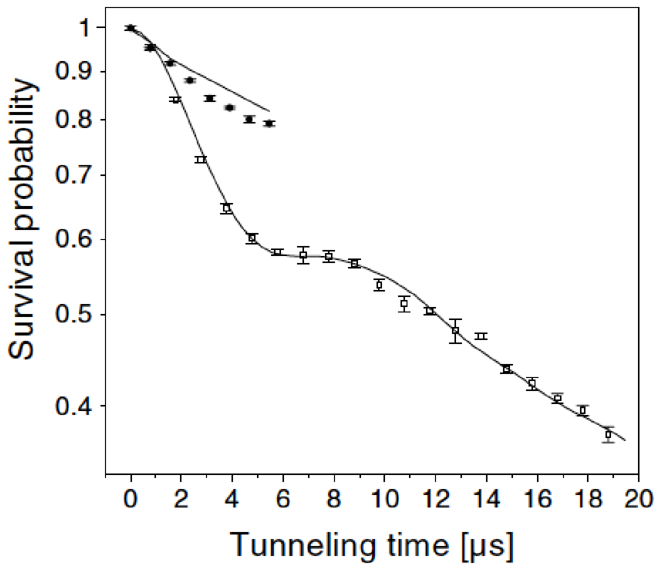
$$\mu_{e+} = -\mu_e$$

## Quantum Zeno and Anti-Zeno effect

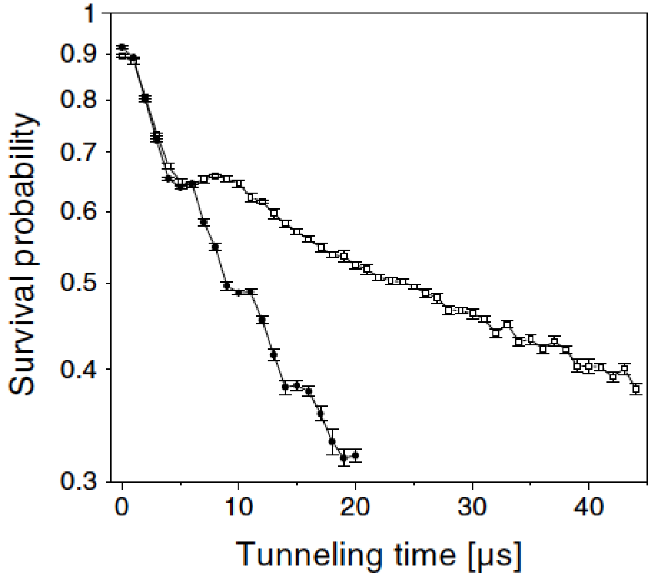
Nonexponential decay in early time. Decay rate very slow in very early time, then accelerates and finally settles to an approximate exponential decay.

Modification of decay rate by the energy spread incurred by the measurement process (result of energy-time uncertainty) and the distribution of states to which the decaying state is coupled.

Quantum Zeno effect




Quantum anti-Zeno effect




Hollow squares →uninterrupted sequence
Solid circles→ Measurement every 1 μs

Hollow squares →uninterrupted sequence
Solid circles→ Measurement every 5 μs



M. C. Fischer et al., Phys. Rev. Lett. **87**, 040402 (2001)

# Formalism for quantum anti-Zeno effect

Consider a quantum system interacting with a reservoir and the reservoir is performing the measurements on the eigenvalue of the quantum system.

Hamiltonian $H = H_0 + V$

|e> is eigenvector of $H_0$ with eigenvalue $\hbar\omega_a = \langle e|H|e\rangle$

|j> describes other eigenvectors of $H_0$ with eigenvalues $\hbar\omega_j$

$V$ describes interaction of |e> with the other states of the reservior.

Wave function of the system is

$$|\psi(t) = \alpha(t)e^{-i\omega_a t} + \sum_j \beta_j e^{-i\omega_j t}|j >$$

Measurements performed at intervals of τ,

Then Survival Probability of |e> after time t (t=nτ)

is = $|\alpha(\tau)|^{2n} \approx \exp(-\lambda_m t)$; $\lambda_m$ is the modified decay rate due to the measurements.

For the case of Quantum Anti-Zeno effect

$$\lambda_m \propto \nu\omega_R^{\eta-1}$$

Where $\nu = 1/\tau$ and $\omega_R$ is frequency of the reservoir.

For EC decay in an atom, electrons performing measurements.

Let $(\Delta E)_{comp}$ be the increase of kinetic energy of the electrons due to compressing atom.

Fractional increase of EC decay rate $\frac{\Delta\lambda}{\lambda} \sim \frac{(\Delta E)_{comp}}{E_{decay}}$; $E_{decay}$ is nuclear decay energy.

## Implanted number of $^{110}$Sn ions

Let C be the number of $^{110}$Sn ions implanted per sec. λ be the decay rate of $^{110}$Sn. $N_1(t)$ be the number of $^{110}$Sn ions present in the catcher foil at the instant t. Then

$$\frac{dN_1(t)}{dt} = C - \lambda N_1(t)$$

$$\frac{dN_1(t)}{dt} + \lambda N_1(t) = C$$

Solution: $N_1(t) = \frac{C}{\lambda}\left(1 - e^{-\lambda t}\right)$

For C=$10^7$ ions/sec $\lambda = \frac{ln2}{4.15 \times 3600}$ sec$^{-1}$, after t= 2 hours

$N_1(t = 2\ hours) = 6.12 \times 10^{10}$ number of $^{110}$Sn ions in the catcher foil.

## Number of $^{110}$In(isomeric) produced during implantation run

$^{110}$Sn decays by electron capture to the isomeric state of $^{110}$In with a half-life =69.1 minutes and it decays by positron emission and EC to $^{110}$Cd.

Let at the instant t, $N_1(t)$ be the number of implanted $^{110}$Sn ions and N(t) be the number of $^{110}$In(isomeric) ions present. Let $\lambda_1$ and $\lambda_2$ be the decay rates of $^{110}$Sn and $^{110}$In respectively and C the number of implanted $^{110}$Sn ions per sec. Then

$$\frac{dN(t)}{dt} = \lambda_1 N_1(t) - \lambda_2 N\,(t)$$

During the implantation run, the equation is

$$\frac{dN(t)}{dt} + \lambda_2 N(t) = \lambda_1 N_1(t) = C\left(1 - e^{-\lambda_1 t}\right)$$

Solution: $N(t) = \frac{C}{\lambda_2 - \lambda_1}\left(1 - e^{-\lambda_1 t}\right)$

After 2 hour implantation,
we get $N(t = 2\ hours) = 2.34 \times 10^{10}$ $^{110}$In ions.

## Number of $^{110}$In ions produced after the completion of 2 hour implantation run

After 2 hour implantation run, the solution is

$$N(t > 2\ hours) = \frac{(6.12 \times 10^{10}) \times \lambda_1}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + (1.956 \times 10^{10}) \times e^{-\lambda_2 t}$$

Number of 280 keV γ-rays emitted per sec from the decay of $^{110}$Sn at the instant t

$$N_\gamma(280) = \lambda_1 \times (6.12 \times 10^{10}) \times e^{-\lambda_1 t}$$

Number of 657 keV γ-rays emitted per sec from the decay of $^{110}$In at the instant t

$$N_\gamma(657) = \lambda_2\left[2.349 \times 10^{10} e^{-\lambda_1 t} + 1.956 \times 10^{10} e^{-\lambda_2 t}\right]$$

Then $\frac{N_\gamma(657)}{N_\gamma(280)} \approx 1.38$ (almost independent of time).

Considering the photopeak efficiency of HPGe detector (80% efficiency)

$$\frac{N_\gamma(657)}{N_\gamma(280)} \approx 0.79$$

Considering positron emission branching ratio of $^{110}$In, the number of 511 keV γ-ray photons going in the general direction of HPGe detector as $N_\gamma(511)$,

$$\frac{N_\gamma(511)}{N_\gamma(657)} \approx 0.71$$

# WIEN2k Density Functional Code

P. Blaha et al., J. Chem. Phys. **152**, 074101 (2020)

Kohn and Sham method ⇛Interacting electrons mapped into a non-interacting system of quasi-particles.

$$E_{Total} = T_s + E_{Ne} + E_{Hatree} + E_{xc} + E_{NN}$$

Electrons move independently in Hatree potential $V_H$ determined from the electron density.

$E_{xc}$ is exchange correlation term coming from Pauli exclusion principle.

Kohn and Sham equation is :

$$\left(-\frac{1}{2}\nabla^2 + V_{KS}(\boldsymbol{r})\right)\psi_i(\boldsymbol{r}) = \varepsilon_i\psi_i(\boldsymbol{r})$$

Where $V_{KS}(\boldsymbol{r}) = V_{Ne}(\boldsymbol{r}) + V_{Hatree}(\boldsymbol{r}) + V_{xc}(\boldsymbol{r})$

$V_{xc}$ (**r**) taken from Perdew, Burke, Ernzerhof (PBE 96) potential.

Basis function taken as :

Inside Muffintin sphere around a nucleus

$$\varphi_{K_n}(\boldsymbol{r}) = \sum_{l,m}\left[A_{l,m,K_n} u_l(r, E_l) + B_{l,m,K_n} \dot{u}_l\,(r, E_l)\right] Y_{l,m}(\boldsymbol{r})$$

In the interstitial region

$\varphi_{K_n}(\boldsymbol{r}) = \frac{1}{\sqrt{\omega}} e^{i\boldsymbol{K_n}.\boldsymbol{r}}$ ; Slope and value of $\varphi_{K_n}(\boldsymbol{r})$ matched at the surface of Muffintin sphere.

# WIEN2k code (continued)

KS orbital wave function

$\psi_{K_n} = \sum_i c^i_{K_n} \varphi_{K_n}(\boldsymbol{r})$

Inside Muffintin sphere

$$V_{KS}(\boldsymbol{r}) = \sum_{l,m} V^{KS}_{l,m}(r) Y_{l,m}(r)$$

In the interstitial space

$$V_{KS}(\boldsymbol{r}) = \sum_{K} V^{KS}_{l,m}(\mathbf{r}) e^{i\mathbf{K}.\mathbf{r}}$$

Similarly,

$$\rho(\boldsymbol{r}) = \sum_{l,m} \rho_{LM}(r) Y_{l,m}(\mathbf{r})$$

$$\rho(\mathbf{r}) = \sum_{K} \rho_{K}(\mathbf{r}) e^{i\mathbf{K}.\mathbf{r}}$$