Positron Annihilation Lifetime Spectroscopy (PALS)

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Introduction

- General idea of the process.

Using positrons, one can get defect information.
What is a positron?

- Theoretical prediction (Dirac, 1928).
  - Non-relativistic QM:
    \[ E = \frac{p^2}{2m} \]
    \(\Rightarrow\) Schrödinger equation
  - Relativistic QM:
    \[ E = \sqrt{\left(mc^2\right)^2 + \left(pc\right)^2} \]
    \(\Rightarrow\) Dirac equation

It seems that negative energies are possible. This led to the idea of antiparticles.
What is a positron?

- Experimental discovery: C.D. Anderson (1932)
What is a positron?

- It was the first antiparticle found experimentally.
- It is similar to the electron but its charge: \( q = e \)
- An electron and a positron annihilate producing two photons (511 keV each).
- Pair production:
  High energetic photons interacting with matter might create a positron electron pair.
How can we obtain it?

- Beta + decay: converts a proton into a neutron, a positron and an electron neutrino.

  \[ p^+ \rightarrow n + e^+ + \nu_e \]

- It can only happen within matter because this process needs energy due to the neutron's mass being greater than the proton's.
How can we obtain it?

- **Na decay scheme**

  - High e+ yield (90.4%).
  - Half-life = 2.6 years.
  - The **1.274 MeV photon** is essential for PALS.
  - Broad energy distribution (from 0.1 eV to 540 KeV aprox.).
How can we obtain it?

**NEPOMUC**

- First a neutron beam hits the Cd(113) sample creating Cd(114) in an excited state.
- Cd(114) decays to its ground state emitting photons.
- This photons create positrons by pair production in Pt.
- The resulting beam is **mono-energetic** and has the world's highest intensity for a mono-energetic positron beam.
How can we obtain it?

- Comparison between the two beams.
PALS: Interaction with solids

- We measure the time between the positron's creation and its annihilation.
- This time provides information about the type of defects present in the sample.

\[
\begin{align*}
\text{t1} & \rightarrow 0.5 \text{ ns} \\
\text{t2} & \rightarrow 2 \text{ ns}
\end{align*}
\]
PALS: Interaction with solids

- PALS' objective is to detect defects in solids, so understanding the interaction between positrons and matter is essential.
Thermalization

- Positrons lose part of their initial kinetic energy due to interactions with the lattice and the electrons.

The implantation profile shows the fraction of positrons that penetrate to a depth $z$.

- Thermalization lasts a few ps ($10^{-12}$ s).
Diffusion

- The positrons start to behave as charged particles inside a lattice: **Bloch states**.

The probability density in interstitial regions is higher.
- The positrons may be trapped in the lattice's defects, this allows us to detect these defects.
- The trapping rate $k$ (number of positrons trapped per time unit) links Bloch states (diffusion) with localized states (trapping).
Trapping

• Relation between the trapping rate and the defect concentration.

\[ k = \mu C \]

C is the defect concentration

\( \mu \) is the positron trapping coefficient (it depends on the type of defect).
Mathematical Model

- Following this model, we will obtain k through PALS technique.
Mathematical Model

• Differential equation for the model.

\[ \frac{dn_b(t)}{dt} = -(\lambda_b + \kappa_d)n_b(t), \]
\[ \frac{dn_d(t)}{dt} = -\lambda_d n_d(t) + \kappa_d n_b(t). \]

\(Nb= \# \text{e+ in the bulk. } Nd= \# \text{e+ in the defect. } \Lambda_b= \text{annihilation rate in the bulk. } \Lambda_d= \text{annihilation rate in the defect. } K_d= \text{trapping rate of the defect.} \)

Starting conditions: \(nb(0)=N ; nd(0)=0.\)
Mathematical Model

• Solution of the equation: 

\[ D(t) = \frac{(nb + nd)}{N} \]

\[ D(t) = I_1 \exp\left(-\frac{t}{\tau_1}\right) + I_2 \exp\left(-\frac{t}{\tau_2}\right), \]

with the abbreviations

\[ \tau_1 = \frac{1}{\lambda_b + \kappa_d}, \quad \tau_2 = \frac{1}{\lambda_d}, \]

\[ I_1 = 1 - I_2, \quad I_2 = \frac{\kappa_d}{\lambda_b - \lambda_d + \kappa_d}. \]

This model can be generalized to various defects
Mathematical Model

Number of positrons in the sample (k defects):

\[ D(t) = \sum_{i=1}^{k+1} I_i \exp \left( -\frac{t}{\tau_i} \right) \]

What we measure in PALS is:

\[ N(t) = \left| \frac{dD(t)}{dt} \right| \quad N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp \left( -\frac{t}{\tau_i} \right) \]
PALS: Experimental Results

- The slopes in the logarithmic plot are the different positrons lifetimes.

\[ N(t) = \sum_{i=1}^{k+1} \frac{I_i}{\tau_i} \exp \left( -\frac{t}{\tau_i} \right) \]
PALS: Experimental Results

• Important relation

\[ \kappa_d = \mu C = I_2 \left( \frac{1}{\tau_1} - \frac{1}{\tau_2} \right) = \frac{I_2}{I_1} \left( \frac{1}{\tau_b} - \frac{1}{\tau_d} \right) \]
PALS: Experimental Results

- Experimental procedure.
  
  Sample A: known type of defect
  
  Sample B: unknown type of defect

  \[ \mu_B = \mu_A \]

  Same type of defect
PALS: Experimental Results

- Experimental scheme.
PALS: Experimental Results

- Sample-source sandwich arrangement.
PLEPS: Pulsed Low Energy Positron beam System

Fig. 4. Example of a lifetime spectrum from the first test-experiments. The sample was a hardened epoxy resin (EEW190). Within a measurement time of 400 s, $3 \times 10^6$ events were accumulated.
PLEPS: Pulsed Low Energy Positron beam System

**Fig. 5.** Positron lifetime spectrum of p-doped SiC at an incident positron energy of $E = 16$ keV. The time resolution is 240 ps (FWHM), the peak to background ratio is $5.7 \times 10^3:1$.

**Fig. 6.** Positron lifetime spectrum of a metallic glass at an incident positron energy of $E = 16$ keV. This spectrum was taken during the test-experiments in July 2007 with an energy selection in front of the pulsing components.
Vacancy Formation and Migration

- Study of vacancy formation and migration using PALS.

\[ \bar{\tau} = \sum_{k=1}^{k+1} I_k \tau_k \]

\[ \sigma C_V = \frac{\bar{\tau} - \tau_f}{\tau_f (\tau_1 - \bar{\tau})} = I_1 \left( \frac{1}{\tau_0} - \frac{1}{\tau_1} \right) \]

\[ = \sigma \exp \left( \frac{S_V^F}{k_B} \right) \exp \left( -\frac{H_V^F}{k_B T} \right) \]

The s-shape curve is due to the thermal vacancy formation

\[
Fe_{61}Al_{39} \quad Fe_{63}Al_{37}
\]

\[ H_V^F = 0.98 \pm 0.07 \text{ eV} \quad \text{or} \quad 1.04 \pm 0.07 \text{ eV} \]
Vacancy Formation and Migration

- Study of vacancy formation through rapid cooling.

\[ \sigma C_V = \frac{\bar{\tau} - \tau_f}{\tau_f(\tau_1 - \bar{\tau})} = I_1\left(\frac{1}{\tau_0} - \frac{1}{\tau_1}\right) \]

\[ = \sigma \exp\left(\frac{S_V^F}{k_B}\right) \exp\left(-\frac{H_V^F}{k_B T}\right) \]

**FIG. 2.** Mean positron lifetime \( \bar{\tau} \) measured on \( \text{Fe}_{61}\text{Al}_{39} \) during equilibration after rapid cooling from the temperature \( T_i = 770 \text{ K} \) to the final temperature \( T_f = 673 \text{ K} \) (\( \triangle \)), 654 K (\( \square \)), 638 K (\( \nabla \)), or 623 K (\( \circ \)).
Vacancy Formation and Migration

- Trapping coefficient variation as a function of temperature.

\[ \Delta C_V(t) = C_V(t) - C_f = (C_i - C_f) \exp\left(-\frac{t}{t_E}\right) \]

\[ t_E^{-1} = \xi \nu_0 \frac{Z}{N} \exp\left(\frac{S_V^M}{k_B}\right) \exp\left(-\frac{H_V^M}{k_B T}\right) \]

\[ F e_{61} A l_{39} \]

\[ H_V^M = 1.7 \pm 0.2 \text{ eV} \]
Vacancy Identification

- Identification of A- and B-Site Cation Vacancy Defects in Perovskite Oxide Thin Films.

We can see the variation in the vacancy concentration as a function of the laser fluence.
References

- **Positron Annihilation in Semiconductors.** R.Krause-Rehberg, H.S.Leipner.
- Wikipedia.
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