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How to analyze condensed matter with Positronium

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XXXI° ciclo

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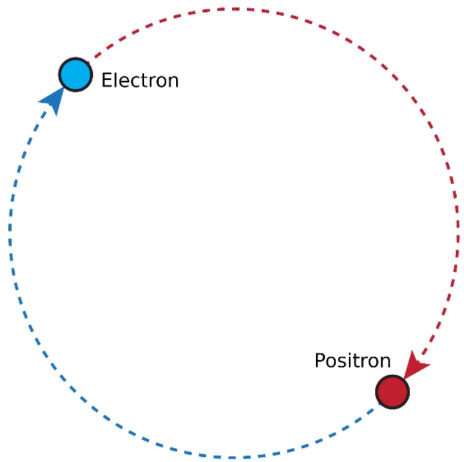
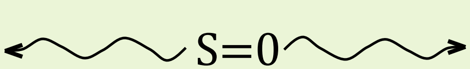
What is Positronium?

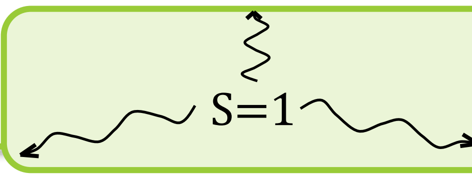
- The simplest bound system composed of matter and antimatter
- Its non relativistic description formally resembles the hydrogen atom
- Its mean lifetime is very small and depends on the spin configuration





$$p\text{-Ps} = \frac{1}{\sqrt{2}}\left(|\downarrow\uparrow\rangle - |\uparrow\downarrow\rangle\right)$$

$$\tau_{para} = 0.125\ ns$$
$$\lambda_{2\gamma} = 8\ GHz$$

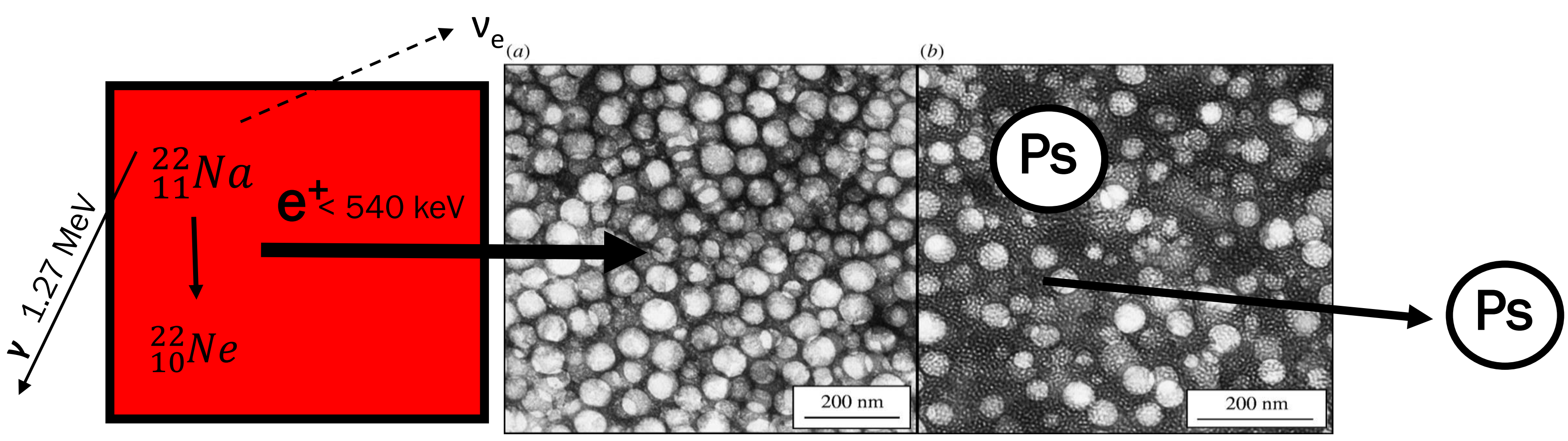


$$o\text{-Ps} = \begin{cases} |\uparrow\uparrow\rangle \\ \frac{1}{\sqrt{2}}\left(|\downarrow\uparrow\rangle + |\uparrow\downarrow\rangle\right) \\ |\downarrow\downarrow\rangle \end{cases}$$

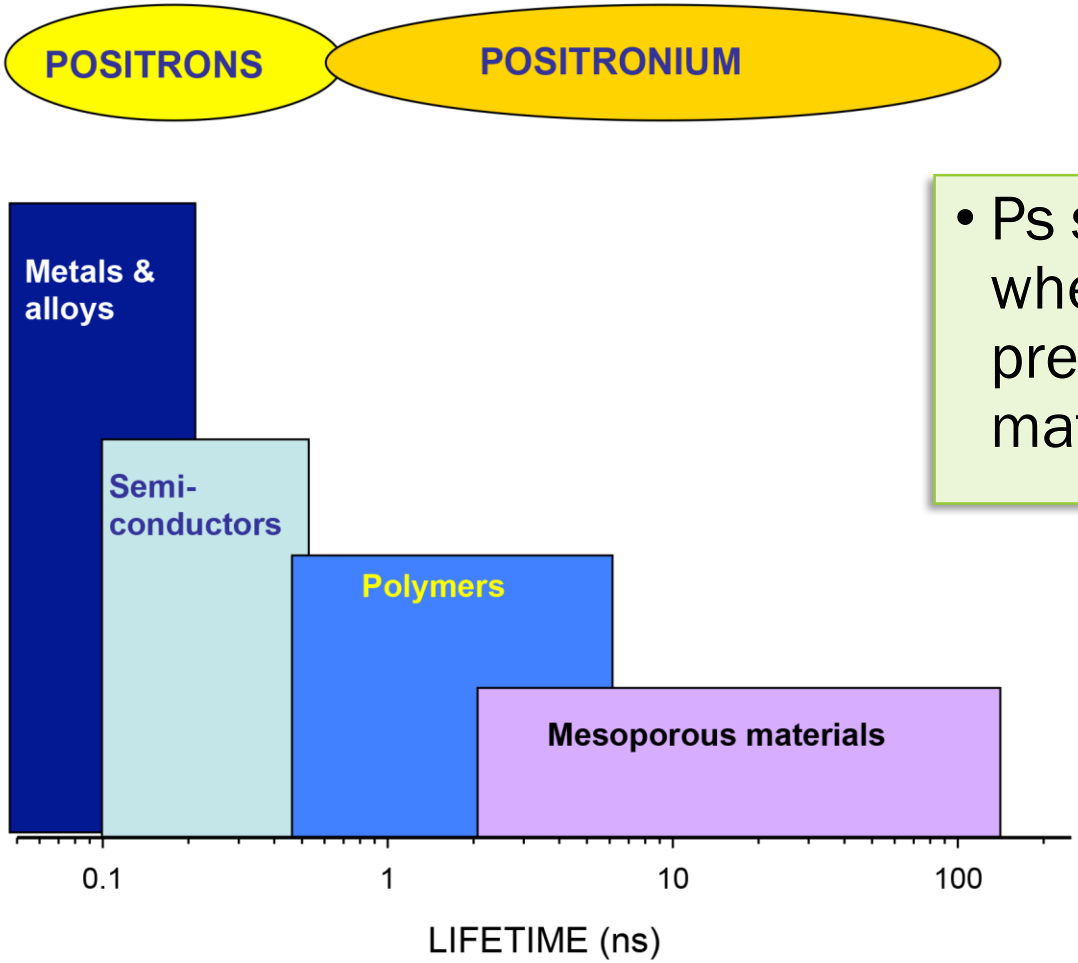
$$\tau_{ortho} = 142\ ns$$
$$\lambda_{3\gamma} = 7\ MHz$$

Where is Ps formed?

- Implanting positrons, usually from a radioactive source, into different solid materials (e.g. amorphous-SiO_2 in the AEgIS experiment at CERN)
- Before annihilation, positrons can thermalize and eventually form Ps in **bulk**
- Ps can escape the material (backscattering or transmission) or **it remains trapped inside** until its annihilation



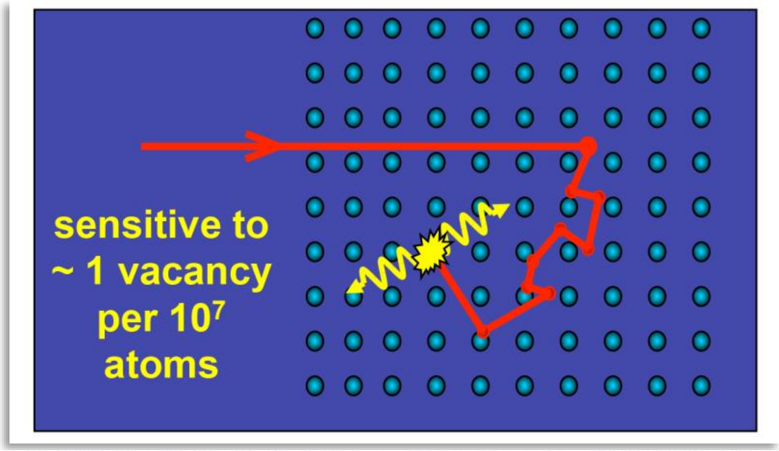
Where is Ps formed?

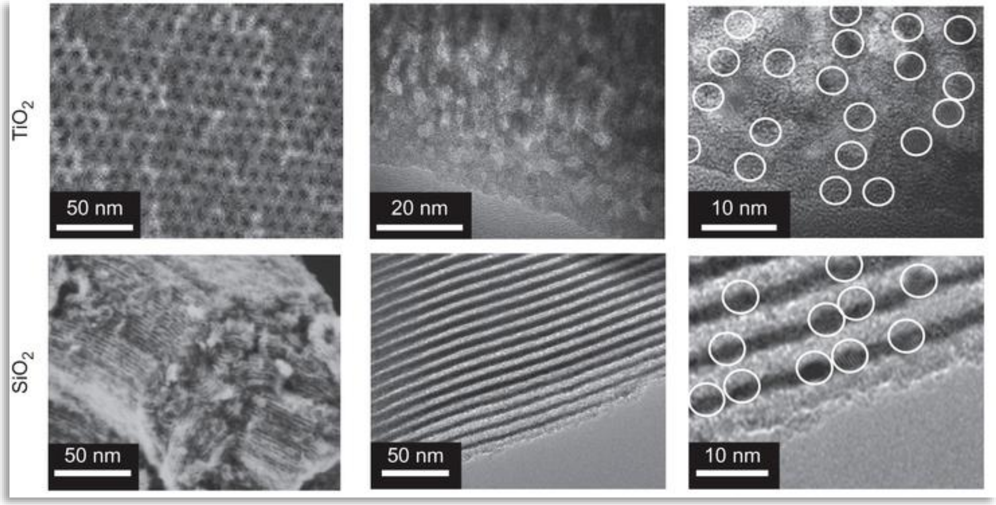


- Ps signature is evident when free spaces are present inside the material

Just porous materials?

Ps presence can be seen in a vast range of open-volume point defects:
from a few ångström to tens of nanometers!



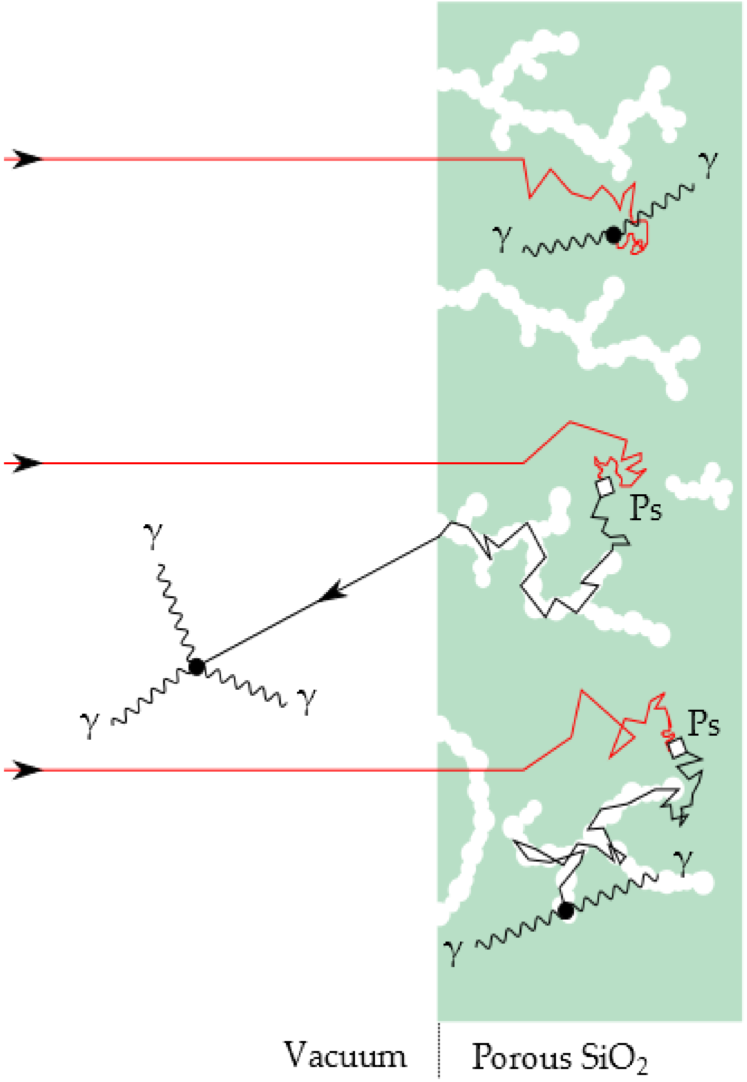


- Small defects like monovacancies, small vacancy clusters, dislocations

- Extended traps like voids, bubbles, precipitates, nanoporous and mesoporous materials

How long does Ps live?

- e+ annihilation in bulk $\tau \approx 0.3\,ns$
- p-Ps formation in pores $\tau \approx 0.1\,ns$
- o-Ps formation in pores ⟶ longer lifetime
  ⟶ it can reach the outer of the material
  $\tau \approx 100\,ns$
- **Pick-off** process:
  the positron annihilates with an external electron, different from the one to which is bound in Ps
  $\tau \approx 2\,ns$



Anihilation in matter: basic concepts

- **Pickoff** annihilation depends on the **electron density** of the material

$$\lambda_{pickoff} \overset{IPA}{\Longrightarrow} \pi r_0^2 c \iiint |\psi_{Ps}\psi(\vec{r}_+,\vec{r}_-)|^2 n(\vec{r})\gamma(\vec{r})\delta^3(\vec{r}_+ - \vec{r})d^3\vec{r}_+ d^3\vec{r}_- d^3\vec{r}$$

- **Intrinsic** annihilation depends on the **contact density**: the probability of finding the positron at the electron position inside Ps

$$k_r \overset{IPA}{\Longrightarrow} \frac{1}{k_0} \iiint |\psi_{Ps}(\vec{r}_+,\vec{r}_-)|^2 \delta^3(\vec{r}_+ - \vec{r}_-)d^3\vec{r}_+ d^3\vec{r}_-$$

- The total annihilation rate for Ps **in matter** can be written as:

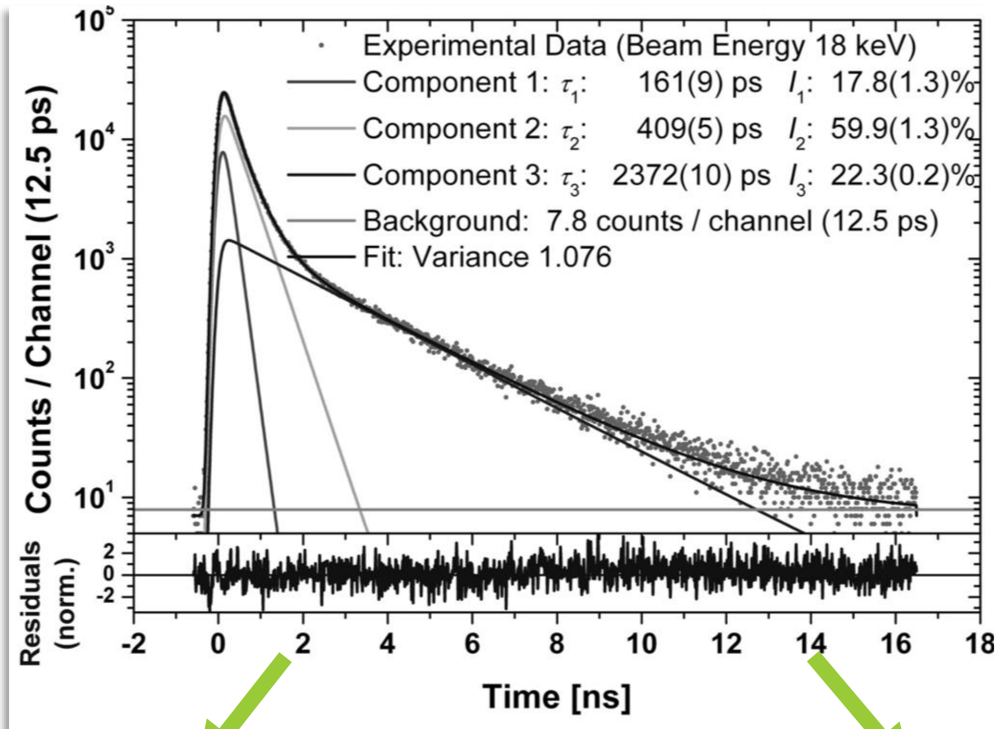
$$\lambda_{para} = \kappa_r \lambda_{2\gamma} + \lambda_{\text{pickoff}}$$

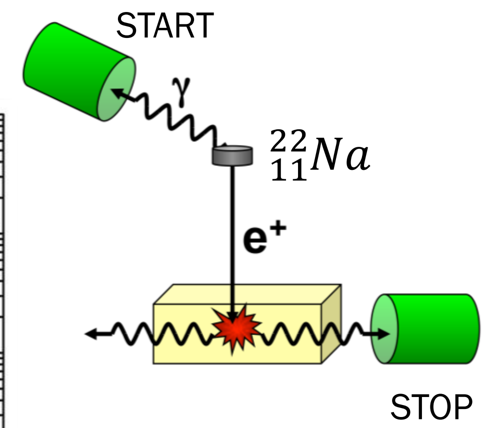
$$\lambda_{ortho} = \kappa_r \lambda_{3\gamma} + \lambda_{\text{pickoff}}$$

Can be measured!

How can be measured?

- Lifetime spectroscopy (PALS)





Without magnetic field:

λ_{pickoff}

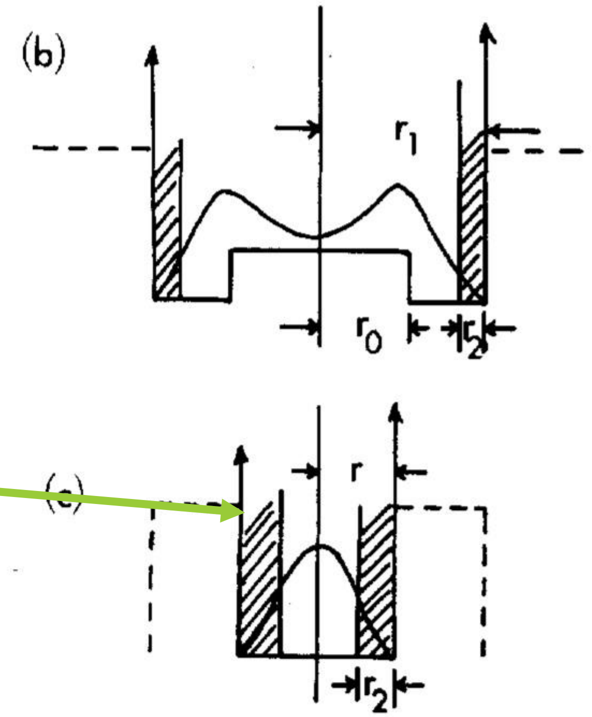
With magnetic field:

κ_r

Theoretical models

- The most famous: **Tao-Eldrup model**

Ps as a **point particle** trapped in an infinite quantum well



An uniform electron density shell $\Delta R = 1.65$Å is taken in the external layer of the cavity to account for Pickoff:

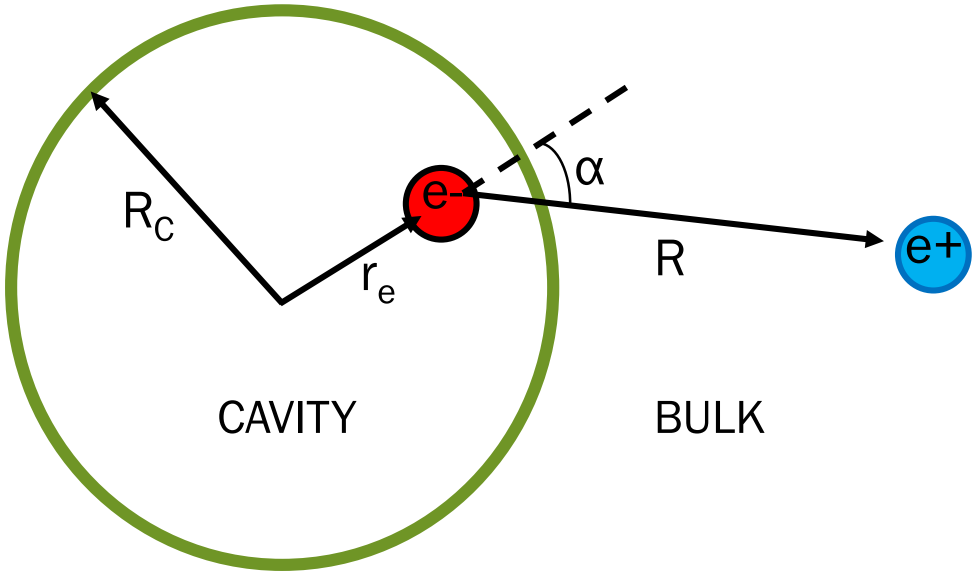
$$\lambda_{pickoff} = (2\ \mathrm{ns}^{-1})\left[1 - \frac{R}{R+\Delta R} + \frac{1}{2\pi}\sin\frac{2\pi R}{R+\Delta R}\right]$$

✓ **Pickoff annihilation rate ↔ Cavity geometry R**

✓ **No information on the contact density** → Needs for new models

Theoretical models

- We have to consider the **internal** structure of the confined Ps to evaluate κ_r → formulation of a new **two-particle** model[1]

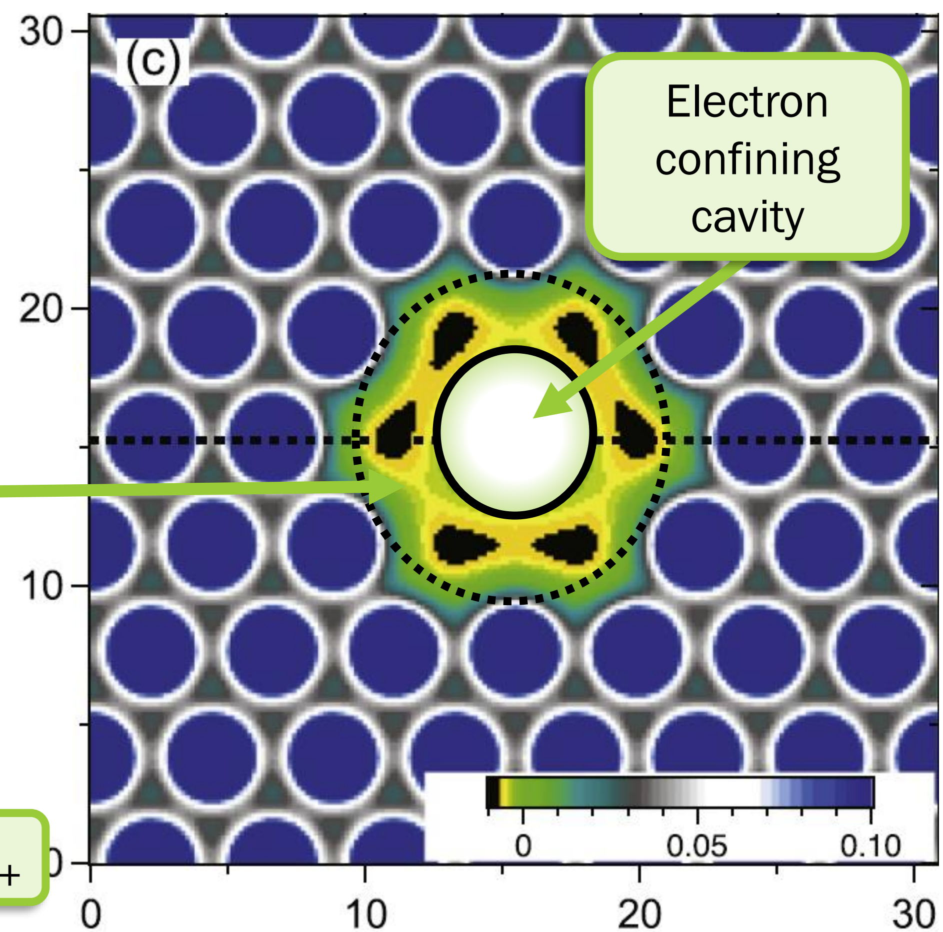


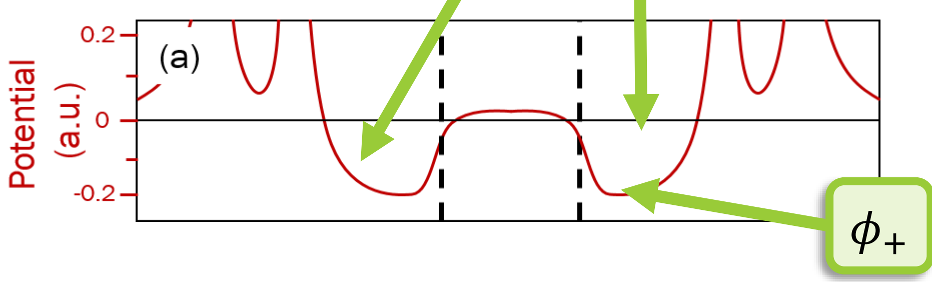
[1] G. Marlotti Tanzi, F. Castelli, and G. Consolati, Phys. Rev. Lett. **116**, 033401 (2016)

Theoretical models

- The electron is confined by the **Pauli exclusion** principle
- The bulk pseudopotential can be related to the **positron work function** ϕ_+ (minimum energy needed to remove the positron from the solid)

Example of DFT calculation in solid He





Theoretical models

- **Two-particle** quantum system where the electron is captured by an infinite potential well and the positron feels the Coulomb attraction of the electron and a bulk pseudopotential

$$H = \left(\frac{\partial^2}{\partial r_e^2} + 2\frac{\partial^2}{\partial R^2} + \frac{2r_e^2 + R^2 + 2r_e R\cos\alpha}{r_e^2 R^2}\frac{\partial^2}{\partial \alpha^2} - 2\cos\alpha\frac{\partial^2}{\partial r_e \partial R} + 2\frac{\sin\alpha}{R}\frac{\partial^2}{\partial r_e \partial \alpha} + 2\frac{\sin\alpha}{r_e}\frac{\partial^2}{\partial R\partial\alpha} + \frac{2}{r_e}\frac{\partial}{\partial r_e} + \frac{4}{R}\frac{\partial}{\partial R} + \frac{2r_e^2\cos\alpha + R^2\cos\alpha + 2r_e R}{r_e^2 R^2 \sin\alpha}\frac{\partial}{\partial\alpha}\right) - \frac{1}{R} + V_{bulk}[\phi_+]$$

Approximated solution in[1]

Better solution?

We need numerical methods!!

[1] G. Marlotti Tanzi, F. Castelli, and G. Consolati, Phys. Rev. Lett. **116**, 033401 (2016)

First Results (work in progress)

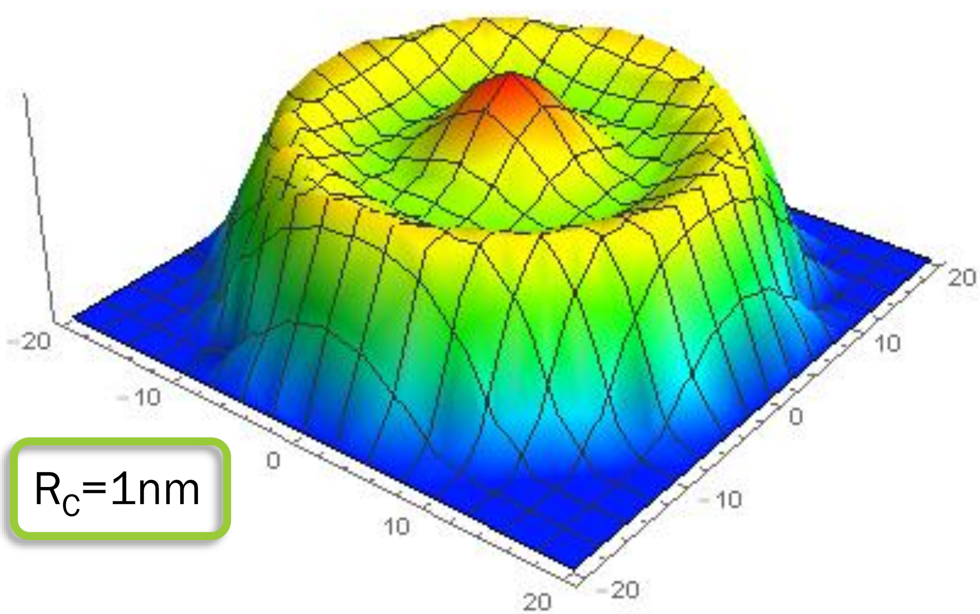
- Ground State : **variational method**

Up to 125 trial parametric wavefunction:

$$\psi_\varepsilon(r_e, R, \alpha) = \sum_{i,j,k} C_{ijk}(R_C^2 - r_e^2)^i (R^j\, e^{-\frac{R}{\varepsilon}})(\cos\alpha^k)$$

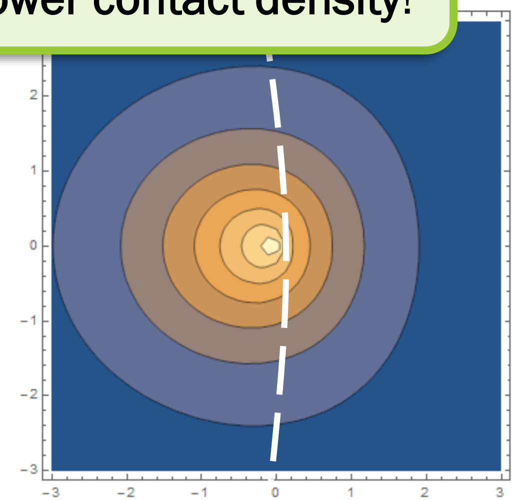
CONFINING — Ps-LIKE — POLARIZATION

Energy minimization with respect to C_{ijk} and ε



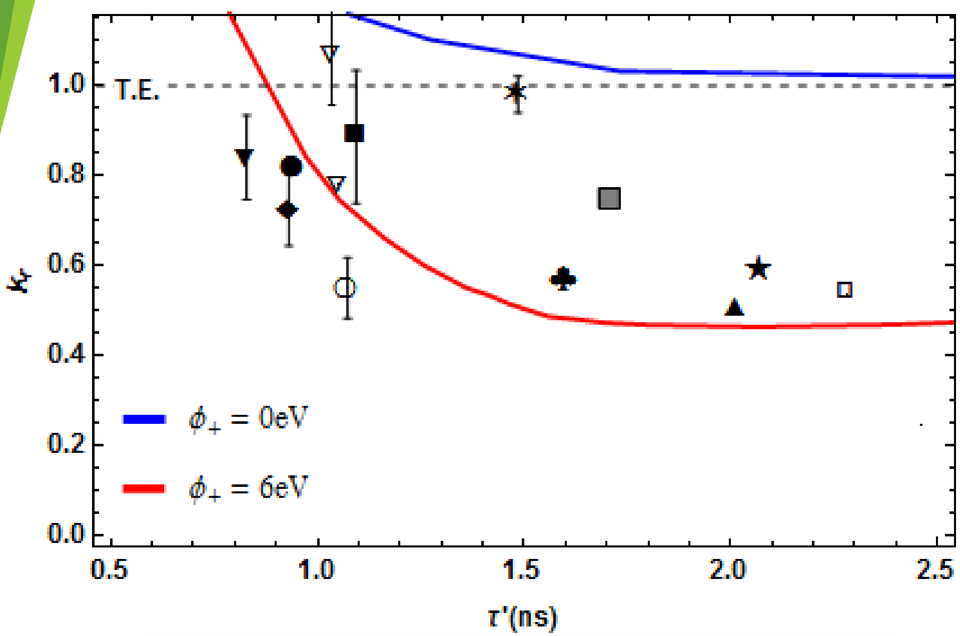
R_C=1nm

Wavefunction Swelling:
Lower contact density!



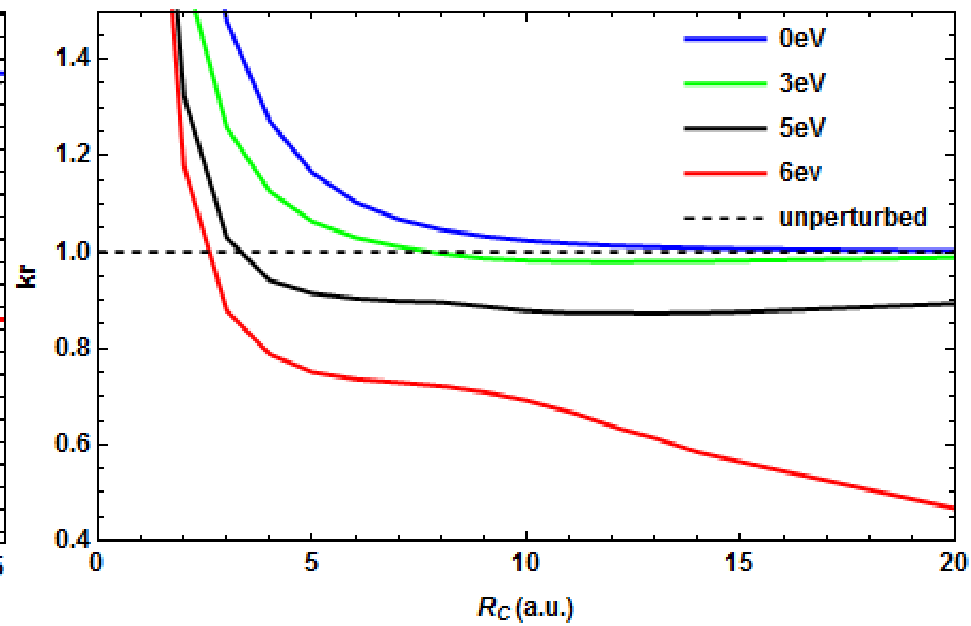
First Results (work in progress)

Contact density as a function of the pickoff lifetime, for different positron work functions

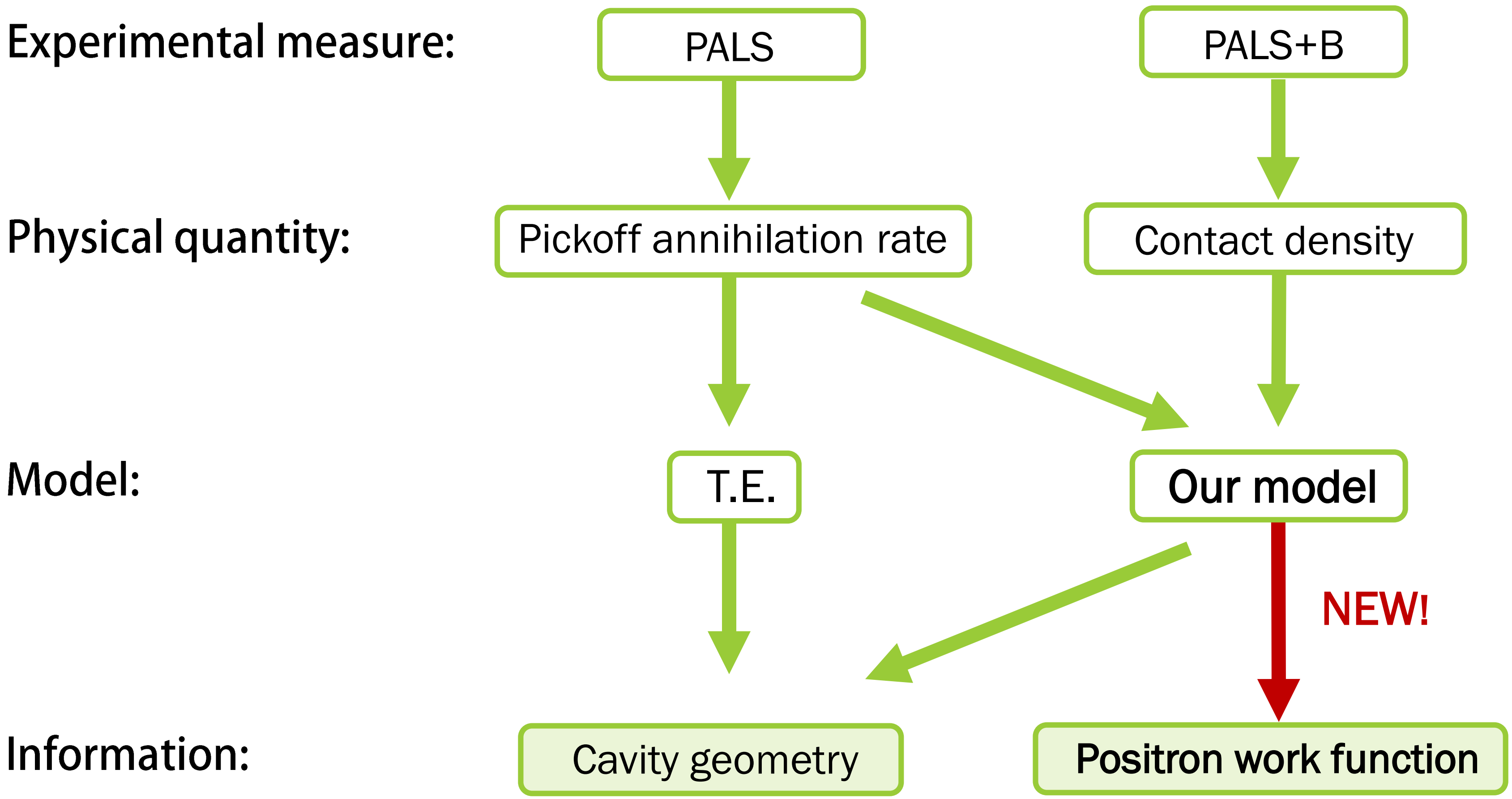


Symbols: known experimental data

Contact density as a function of the cavity radius, for different positron work functions



To recap



Experimental measure:
PALS
PALS+B
Physical quantity:
Pickoff annihilation rate
Contact density
Model:
T.E.
Our model
NEW!
Information:
Cavity geometry
Positron work function

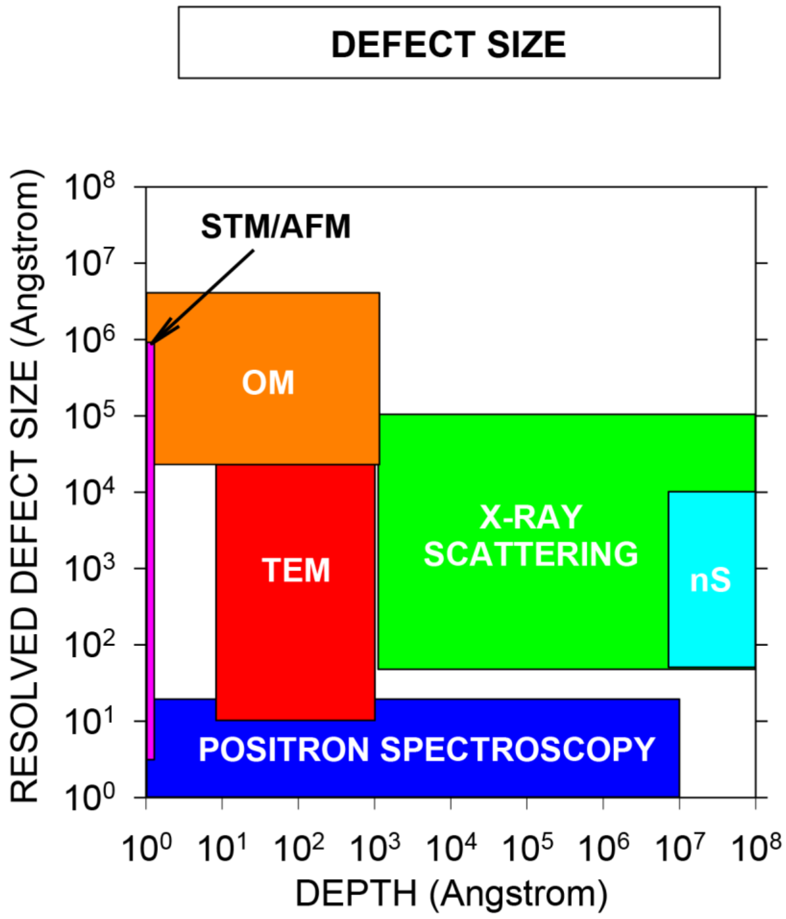
Conclusions

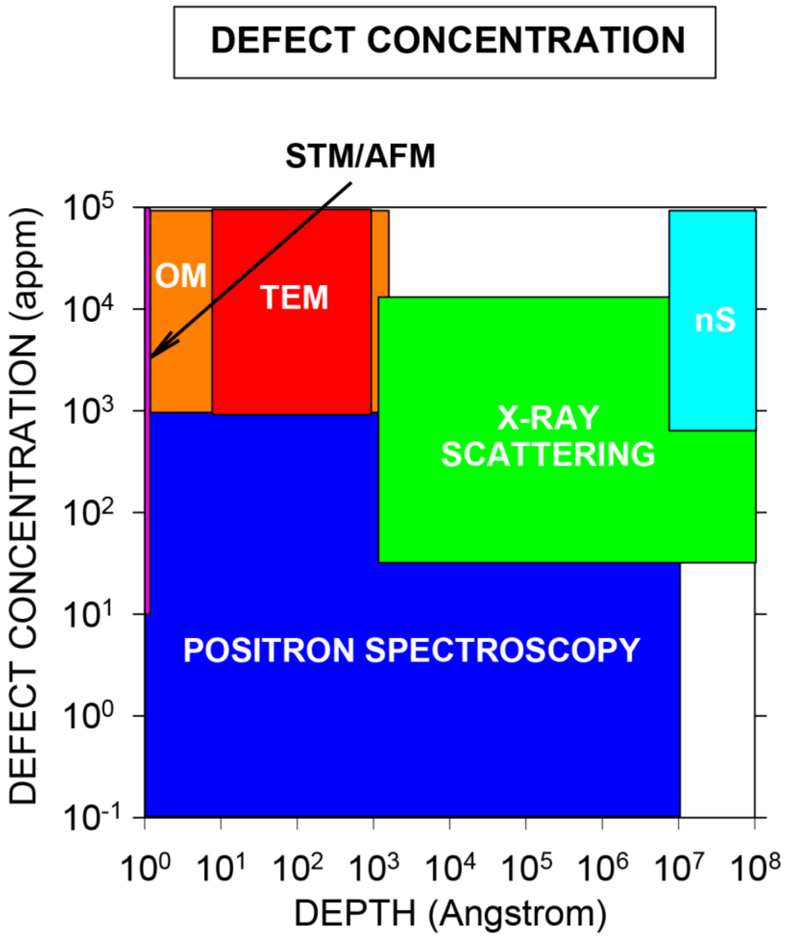
- Ps is a **useful probe** to study condensed matter
- We're studying a model capable of connecting Ps properties (pickoff and contact density) with properties of the surrounding material (work function, pore dimensions)
- Next investigation will regard DFT calculation for pseudopotentials and the analysis of the relation between the material and the positron spin polarization

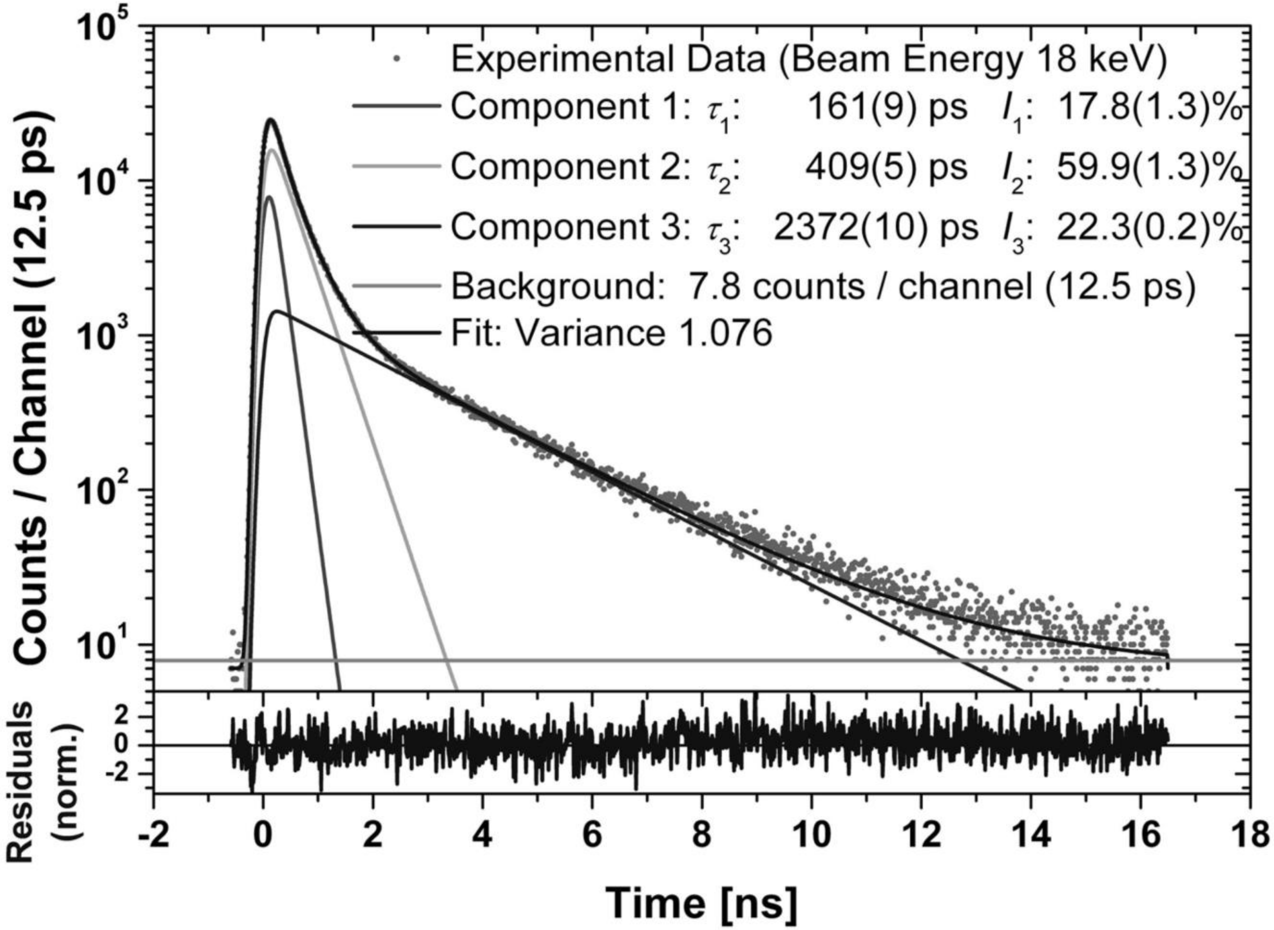
Thank you for your attention

Extra slide

Why PALS?


DEFECT SIZE
STM/AFM
OM
TEM
X-RAY SCATTERING
nS
POSITRON SPECTROSCOPY
RESOLVED DEFECT SIZE (Angstrom)
10^0 10^1 10^2 10^3 10^4 10^5 10^6 10^7 10^8
DEPTH (Angstrom)


DEFECT CONCENTRATION
STM/AFM
OM
TEM
nS
X-RAY SCATTERING
POSITRON SPECTROSCOPY
DEFECT CONCENTRATION (appm)
10^{-1} 10^0 10^1 10^2 10^3 10^4 10^5
10^0 10^1 10^2 10^3 10^4 10^5 10^6 10^7 10^8
DEPTH (Angstrom)



Typical positron lifetime spectrum in an epoxy-based industrial adhesive. The spectrum can be decomposed into three exponentials with three different lifetimes. From the longest lifetime of 2.3 ns the mean size of free volume cavities in the polymer is estimated to be 0.13 nm.

Confining effect

- The confining effect is mostly due to Pauli exclusion principle

Coulombic repulsion without Pauli

Pauli exclusion principle keeps the positron outside the atom

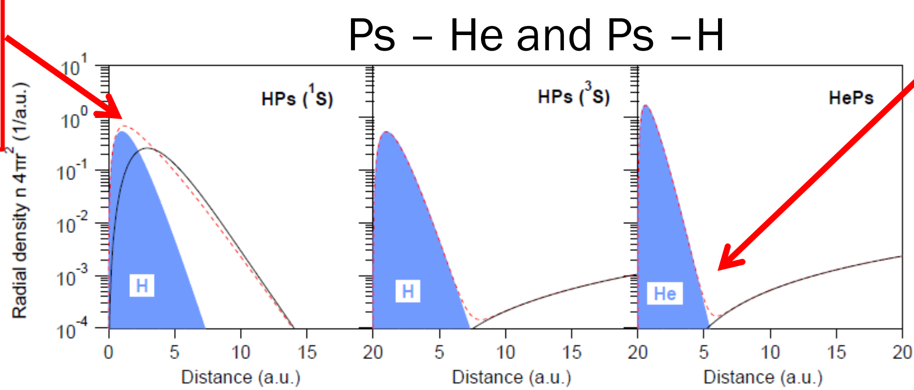


Figure 1. (Color online) Electron and positron densities of ^1S_e-HPs, ^3S_e-HPs, and HePs. The electron densities of the isolated atoms (filled blue curves), and the interacting positron-atom systems (red broken curves), as well as the positron densities (black full curves) are shown.