Ps formation and Ps cooling (II)

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I lecture

• Ps thermalization and cooling by collisions
  Thermalization in gases
  • Thermalization in silica oxide powders
  • Ps emission at low temperature from modified metal surfaces

II lecture

• Porous materials: cooling at room temperature
  • Porous materials: cryogenic cooling
  • oPs pick-off at cryogenic temperature
  • oPs emission from nano-channels in silicon
Production of porous materials

- Deposition by spin-on or deep coating with sacrificial porogen

- Silica based, with tetrahedral basic structure
  Precursors: $TEOS \rightarrow Si(OC_2H_5)_4$, $TES \rightarrow HSi(OC_2H_5)_3$
  $MTES \rightarrow CH_3Si(OC_2H_5)_4$

- Silsesquioxane (SSQ) based: organic-inorganic polymers $(R-SiO_{3/2})$;
  $R = H(HSSQ)$; $R=CH_3$ MSSQ


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oPs is identified by:

- lifetime measurements: oPs lifetime shortened by pick-off mechanism
- Peak to Valley ratio in Doppler measurements: 2gammas/3gammas
Ps in silica based materials is emitted into porosities with an energy of 1-3 eV, time of flight measurements are necessary to know the energy of the Ps emitted after collision with the pore walls.
Thermalization and cooling depends from:

- Size of the porosities
- Energy of oPs
- Number of collisions
- Energy loss for collision
- Chemical termination of the pore walls
- Temperature of the environment

Yield of cooled oPs is influenced by $2\gamma$ annihilation (pick-off and spin exchange)
Ps confinement

Positronium de Broglie wavelength

\[ \lambda = \frac{\hbar}{m_{Ps} v} = \frac{76}{\sqrt{T}} \text{ nm} \]

Ps can remain confined an excite states

Energy levels

\[ E = \frac{\pi^2 \hbar^2}{2m_{Ps}} \left( \frac{n}{a} \right)^2 \]
Ps injected in the cavity is very hot

Models for energy loss $\Delta E$:

a) Classical elastic scattering with ensemble of effective mass $M^*$

b) Phonon scattering

Classical elastic scattering is expected to hold only in large cavities and not to describe the system at low temperature
The times for reaching the thermal energy depends from the size and the temperature of the environment.

Characteristic thermalization times can reach several nanoseconds lowering T, the energy loss decrease decreasing T.

At low T the permanence in excited states can become important!
Pick off: when the e+ in Ps has an overlap with an e- of the walls in an interaction region R

$$\lambda_{pick-off} \propto \frac{v_{Ps}}{L}$$

Spin exchange in presence of unpaired e-

$$\lambda_{spin-conversion} = \frac{v_{Ps}}{L} \frac{\sigma}{C} \mu s^{-1}$$

$$C \text{ paramagnetic centers cm}^{-2}$$

$$\sigma = \text{cross section} \approx 10^{-18} \text{ cm}^{-2}$$

Paramagnetic centers produced by radiation are stable at low T
The pick off is higher for high Ps energy

It is expected:

a) to decrease, decreasing the temperature (in some pore size range)

but also

b) to depend from cooling time: ie. permanence in the excited states

Model for lifetime in pores (due to pick-off): RTE model for T > 300K

RTE : rectangular Tao-Eldrup model
Ps in pores at room T and above

The behavior of Ps lifetime is well described by Rectangular Tao-Eldrup (RTE) quantum mechanical model.

The model can be assumed as benchmark for looking for deviation of experimental data below RT.

Hypothesis of RTE:

a) Ps is considered thermalized with the medium, thermalization time is not considered.

b) Ps samples all excited states given by a Boltzmann distribution.
The annihilation rate \( \lambda_{RTE} \) is weighted in each region with the square of the Ps wavefunction.

Then average on all the oPs the energy levels populated with a probability given by the Boltzmann distribution.

\[
\lambda_T \quad \lambda_A = \frac{\lambda_S - 3\lambda_T}{4}
\]

Spin average rate in a region \( R \) from the surface

\[
\lambda_{RTE} = \lambda_A - \frac{\lambda_S - \lambda_T}{4} \left[ 1 - \frac{2R}{a} + \sum_{i=1}^{\infty} \frac{1}{i\pi} \sin \left( \frac{2i\pi R}{a} \right) \exp \left( -\beta \frac{i^2}{a^2 k_B T} \right) \right]^3
\]

\[
\beta = \hbar^2 \pi^2 / 4m
\]

\( R = 0.18 \text{ nm} \)
PROVE of Thermalized Ps at $T > 300$

\[ \lambda_{RTE} = \lambda_A - \lambda_S - \lambda_T \frac{2R}{a} \left[ 1 - \sum_{i=1}^{\infty} \frac{1}{i\pi} \sin \left( \frac{2i\pi R}{a} \right) \exp \left( -\beta \frac{i^2}{a^2 k_B T} \right) \right]^{3} \]

\[ \beta = \frac{\hbar^2 \pi^2}{4m} \]

Decrease of $\tau$ with $T$

Material: capped TEOS (Tetraethylorthosilicate)

from Gidley et al. PRB60, R5157 (1999)
Experimental evidences at RT

Thermalization at RT

Dependence of Ps cooling from pores decoration

Dependence of Ps cooling from type of connected porosity
Ps emitted by a mesoporous material: at 4 keV, Ps follow a Maxwell Boltzmann distribution.
Ps are emitted at near thermal energy $= \frac{3}{2} kT$. 

From Vallery et al. PRL 90, 203402 (2003)
Dependence from pore surface

\[ E(t) = \coth^2(\beta + \alpha t) \]

\[ \beta = \coth^{-1} \sqrt{\frac{E_0}{E_{th}}} \]

\[ \alpha^* = \frac{2}{LM_s} \sqrt{3k_bT m_{Ps}} \]

From C. He et al. PRB 75, 195404 (2007)

\[ M_{-OH} \equiv M_{-H/-OH} < M_{-CH_3/-OH} < M_{-CH_3} \]

13 amu 37 amu

Size porosities \(~3-3.5\) nm
Dependence from type of channels

HSSQ  \((=\text{CH-SiO}_{3/2})_n\)

MSSQ  \((H-\text{SiO}_{3/2})_n\)

Same pore size \(\sim 3.5\)
But different tortuosity (?)

Silica zeolite: bimodal distribution of pore,
Wormlike mesopores (4 nm) and micropores in zeolite (0.5 nm)

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Cryogenic cooling - phonon scattering

Phonon model: interaction with longitudinal acoustic phonons

Deformation potential

\[ W = E_d \sum_q \left[ \sqrt{\frac{\hbar}{2NM\omega_q}} i q \left( a_q e^{i\vec{q}\cdot\vec{r}} - a_q^+ e^{-i\vec{q}\cdot\vec{r}} \right) \right] \]

\[ E_d = 3.6 \text{ eV} \]

With the first order perturbation theory

\[ P_{k',k} = \left| \frac{i}{\hbar} \int_0^t \sum_q \left< k' \left| E_d \frac{1}{\sqrt{N}} \left[ \sqrt{\frac{\hbar}{2M\omega_q}} i q \left( a_q e^{i\vec{q}\cdot\vec{r}} - a_q^+ e^{-i\vec{q}\cdot\vec{r}} \right) \right] k \right> e^{i \frac{E_{\text{Ps}}(\text{final}) - E_{\text{Ps}}(\text{initial}) + \sum_q \hbar \omega_q \left( n_q^f - n_q \right)}{\hbar} t} \right|^2 \]

And total wavefunction before and after Ps scattering

\[ |k\rangle = |Ps(\vec{r})_k \rangle \Pi_q |n_q\rangle \quad \quad |k'\rangle = |Ps(\vec{r})_{k'} \rangle \Pi_q |n_q^f\rangle \]

It is solved in the linear region of the acoustic branch

\[ \omega_q = v_S q \]

Nagai et al. PRB 2000

PRB 08 Mariazza-Salemi-Brusa

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In an infinite rectangular well, the Ps wavefunction is

\[ |\psi(r)\rangle = \sqrt{\frac{2}{a}} \sqrt{\frac{2}{b}} \sqrt{\frac{2}{c}} \sin(k_x x) \sin(k_y y) \sin(k_z z) \]

The permitted energy levels are

\[ E = \frac{\hbar^2 (k_x^2 + k_y^2 + k_z^2)}{2m} = \frac{\hbar^2 k^2}{2m} \]

Solving for the transition probability \( P_{kk'} \):

Energy conservation

\[ (k^I)^2 - k^2 \pm \frac{2mv_s}{\hbar} q = 0 \]

Momentum conservation

\[ \vec{k}^I - \vec{k} = \vec{q} \]

The maximum admitted variation for momentum is

\[ \Delta k_{\text{max}} = \left| k \right| - \left| k^I \right| \approx \frac{2v_s m}{\hbar} \approx 1.7 \times 10^8 \text{ m}^{-1} \]

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The condition implies that NO transition (at the first order) can occurs if the distance between levels is higher than

$$\Delta E_{\text{max}} = \frac{\hbar^2 \Delta k_{\text{max}}}{2m}$$

the lower accessible level \( n_m \) can be found for each box of dimension \( a \)

And then the minimum energy and the minimum \( T \)

$$E = \frac{\hbar^2 \pi^2}{2m} \left( \frac{n_m}{a} \right)^2$$

$$T = \frac{2}{3k_B} E = \frac{\hbar^2 \pi^2}{3k_B m} \left( \frac{n_m}{a} \right)^2$$
Minimum energy and minimum temperature of a Ps atom confined in a pore of size $a$

\[ E = \frac{\hbar^2 \pi^2}{2m} \left( \frac{n_m}{a} \right)^2 \]

\[ T = \frac{2}{3k_B} E = \frac{\hbar^2 \pi^2}{3k_B m} \left( \frac{n_m}{a} \right)^2 \]

$n_m$ is the lower accessible level

Dashed line: ground level

Continuous line: ground state = minimum level in Nano-channels
Cooling time by phonon scattering

The cooling time can be calculated in an infinite potential well in which there is always a level for the transition.

Distance between levels equal or smaller than \( \Delta k_{\text{max}} = |k| - |k'|_{\text{max}} = \frac{2v_{sm}}{\hbar} \approx 1.7 \times 10^8 \text{ m}^{-1} \)

True for dimension > 20 nm

\[
\frac{dE(t)}{dt} = \Delta E(T) \frac{v_P s}{a} \\
\text{Solving for the transition } kk' \text{ } P_{kk'}
\]

and considering a continuum slowing down the average energy loss is calculated

\[
\langle \Delta E(k) \rangle = \frac{a}{\pi} \int \Delta E P(\Delta E) d\Delta E = \frac{a}{\pi} \int \frac{\hbar^2}{2m} \left( k^2 - k'^2 \right) P_{kk'} dk' dI
\]
\[ \langle \Delta E(k) \rangle = \left| \frac{E_d}{a} \frac{1}{\sqrt{N}} \right|^2 \frac{aL}{2\pi^2} 8mR \pi^2 \frac{2R}{M} \left( 2k - \frac{2v_m m}{h} e^{-\frac{hB(2k+2mv_x)}{k_BT}} - \frac{2k - 2v_m m}{h} e^{-\frac{hB(2k-2mv_x)}{k_BT}} - \frac{2k - 2v_m m}{h} \right) \]

\[ t = \frac{R}{v} \approx 2 \frac{Rm}{\hbar k} \]
Thermalization time vs. pore size

Calculation at 10 K
The obtained results with the phonon scattering quantum model about Ps confining and Ps cooling time can be used for:

a) Discuss the deviation of lifetimes at low T from RTE model

b) To design a porous material to cool Ps at low T
Deviation from RTE model at LT

Mesoporous MSSQ films

≤ 3.2 nm

≤ 2.8 nm

≤ 2.1 nm

Lynn -05

Silica Glass

Krause-Rehberg -07
If we exclude:

i) change in the chemistry of the pore
ii) Preferential trapping of Ps in pore of different dimensions changing T
iii) Different Ps formation and diffusion at different T
iv) Production of unpaired electrons at low T
nano-channel in Si –why?

\[ \Phi = 10-15 \text{ nm} \]
Pore diameter around 10 nm

Si p-type

SEM Top view

SEM Cross view
$F = \left[ 1 + (R_{100} - R_F)(R_F - R_o)^{-1}(P_{100}/P_o) \right]^{-1}$

Si p-type (0.15-0.21 Ohm cm)

oPs diffusion length $L_{Ps} \sim 500$ nm

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nano-channels in Si- how they works?

\[ L_+ \sim 200 \text{ nm}, \text{ increasing decreasing the temperature} \]

Silica volume 10-20 % of Si
Advantage

- Less damage than in silica (paramagnetic centers)
- Possible control of the internal surfaces
  (type of oxide, H termination)
- Less sensitive to charging effects

Drawback at very low T (< 1K)

- $^3P_s$ sticking on the surfaces of the pores (?)
- $e^+$ quantum reflection to the Si/SiO$_2$(?)
  (see Britton et al. PRL 62,2413 (89))

\[
T = \frac{2[k_B T(k_B T - 2\phi_+)]^{1/2}}{k_B T - \phi_+ + [k_B T(k_B T - 2\phi_+)]^{1/2}}.
\]

Transmission in metal

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Cooling of Ps seems feasible

At room temperature Ps emitted with near thermal energies was observed.

At cryogenic temperature we have only evidences in SiO$_2$ powders and in Al covered O of Ps emitted in vacuum with T minor than RT.

Porous materials and new organized structures seems to be promising target for Ps cooling but there is a lack of investigation at low T.

Ps confinement has an important role in defining the final energy of cooled Ps.