Characterization of CPG Materials by Using PATs

Presented

by

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Introduction:

- Preparation of Porous Materials
- Principles of Ps in Porous Glass

Investigation of CPGs By PAT

- Calibration of pore sizes by using o-Ps lifetime
- Simulation of the PALS spectra
- T-dependence Measurments
- Cryo-Condensation Phenomenon

Preparation of Porous Materials

1. Melting

The mixture of CPGs or PVGs

Heat treatment → 500-800 °C

2.Phase separation

rich in Silica Borosilicate and Alkial

Leaching out → Hot acid sol.

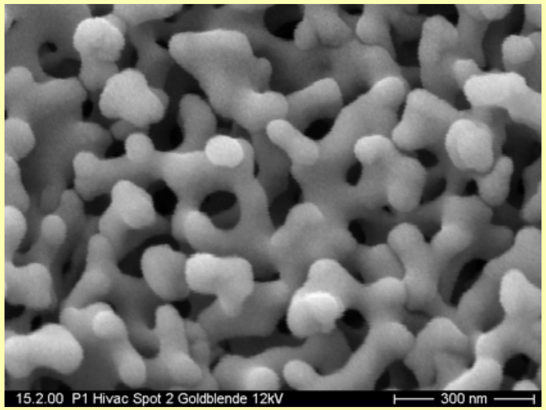
3. Dissolving Borate

Porous Glass Free from the Borate Phase

Heating Above 1200 °C → NaOH

Porous Glass with 96 % SiO_2

glas

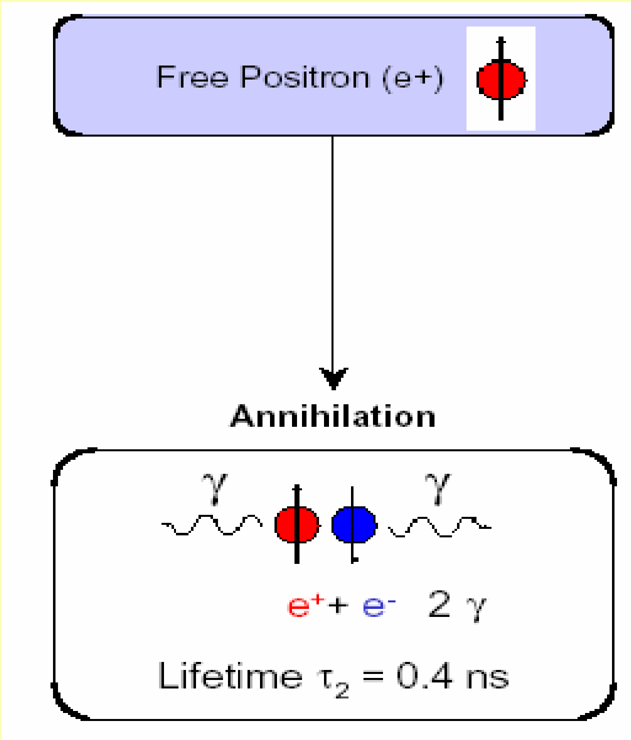


D ------ 1 bis 120 nm

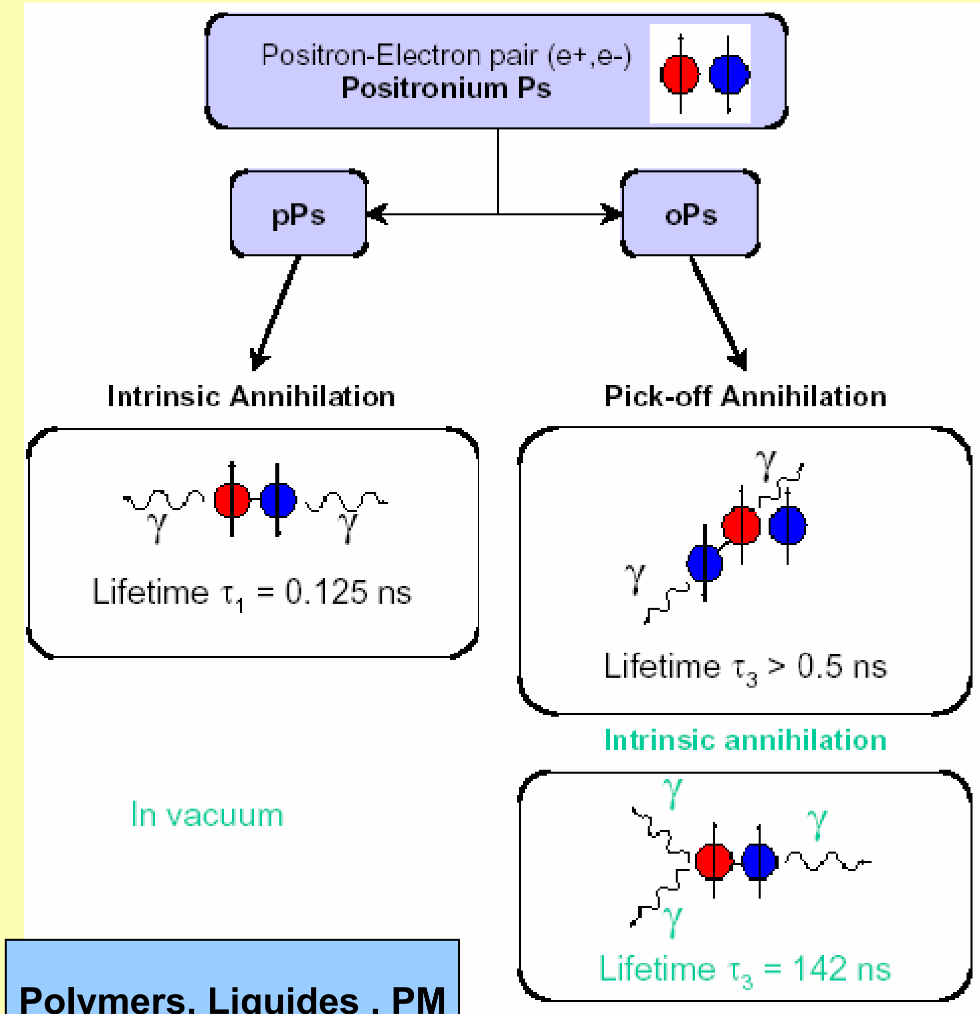
- ***Micropores (d_P < 2 nm)***
  - ***Ultramicropores (d_P < 0.7 nm)***
  - ***Supermicropores (d_p > 0.7 nm < 2 nm)***
- ***Mesopores (d_P = 2 bis 50 nm)***
- ***Macropores (d_P > 50 nm)***

	PVGs	CPGs	MCM-41
Pore size	4-7 nm	4.5 -400 nm	1-10 nm
Pore shape	spherical	Cylinderical spherical	cylindrical
Porosity	50-75 %	28 %	--------
Surface Area	10-350 m^2/g	90-200 m^2/g	---------

Basic Principles of Ps in Porous Glass



Metals, Alloys, Semi-



Polymers, Liquides , PM

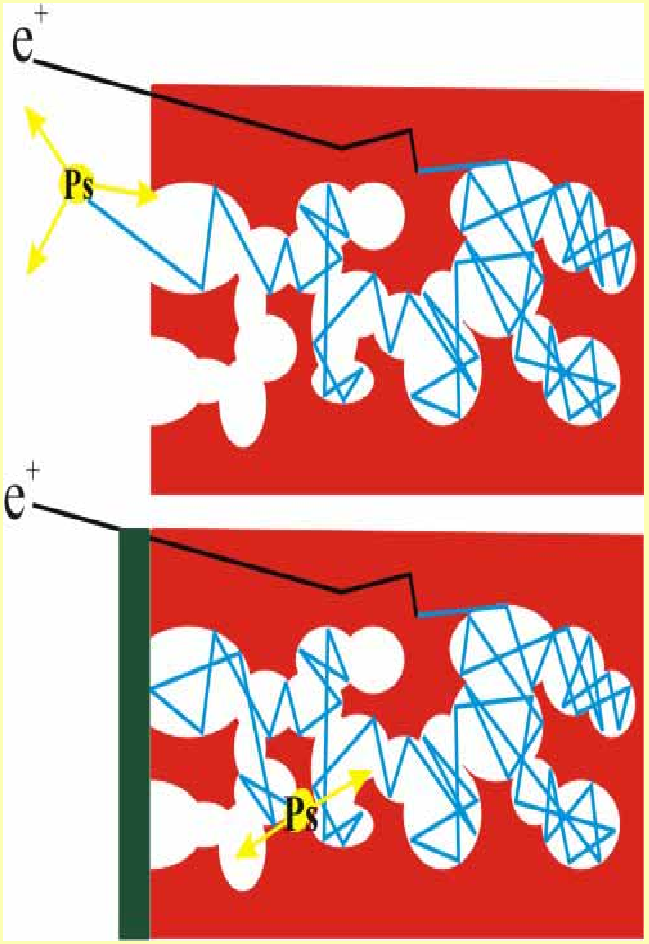
Basic Principles of Ps in Porous Glass

(a) Open Porosity

The travel distance of Ps can be greater than the porous film thickness (usually less than 1 micron in low-k films).

As a result, Ps can easily diffuse out of the film and into the surrounding vacuum.

The observable effect on the Ps lifetime is that most of the Ps annihilates with the vacuum lifetime of 142 ns, a telltale indicator that the pores in the film are interconnected.
A capping layer is required to get the correct Ps lifetime in the interconncted pores.

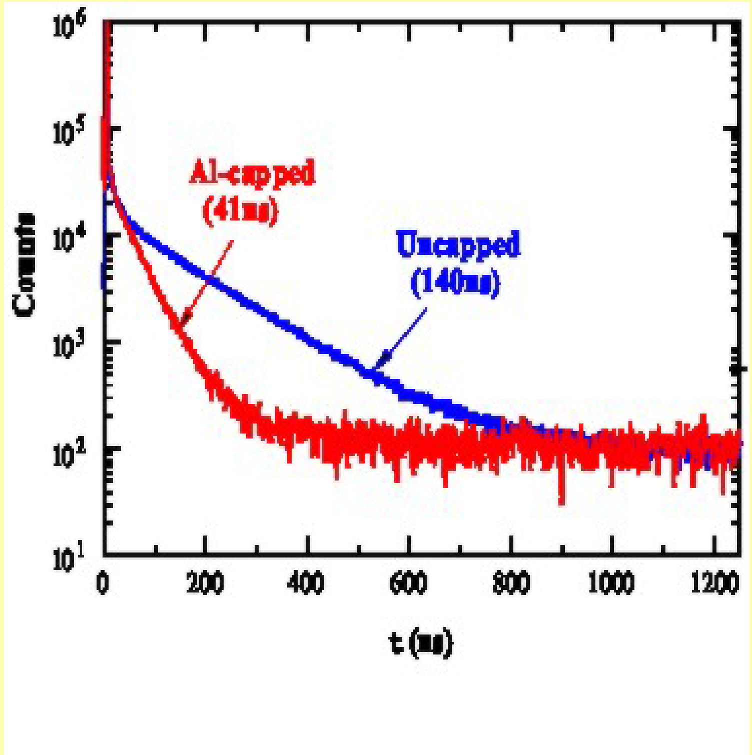


Basic Principles of Ps in Porous Glass

There is a prompt peak characterized by positron and Ps lifetimes less than 0.5 ns (nonporous, bulk samples) .

long lifetime component indicative of copious (35 - 40%) Ps formation in the pores with fitted value of 140 ns, suspiciously indistinguishable from the vacuum value.

It indicates that the pores are so highly interconnected that mobile Ps is able to diffuse out of the film and into the surrounding vacuum system.



$$\lambda_{2\gamma} = 2ns^{-1}W = 2ns^{-1}\left[1 - \frac{r_h}{r_h + \delta r} + \frac{1}{2\pi}\sin\left(\frac{2\pi r_h}{r_h + \delta r}\right)\right]$$

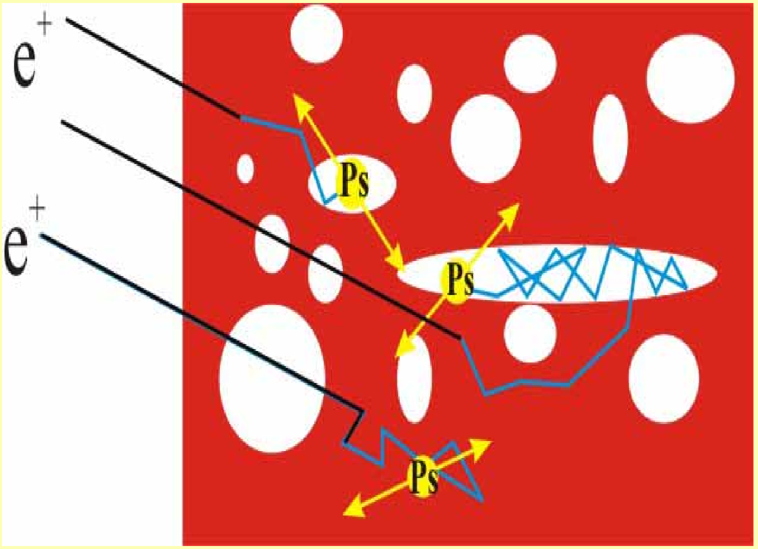
(a) Closed Porosity

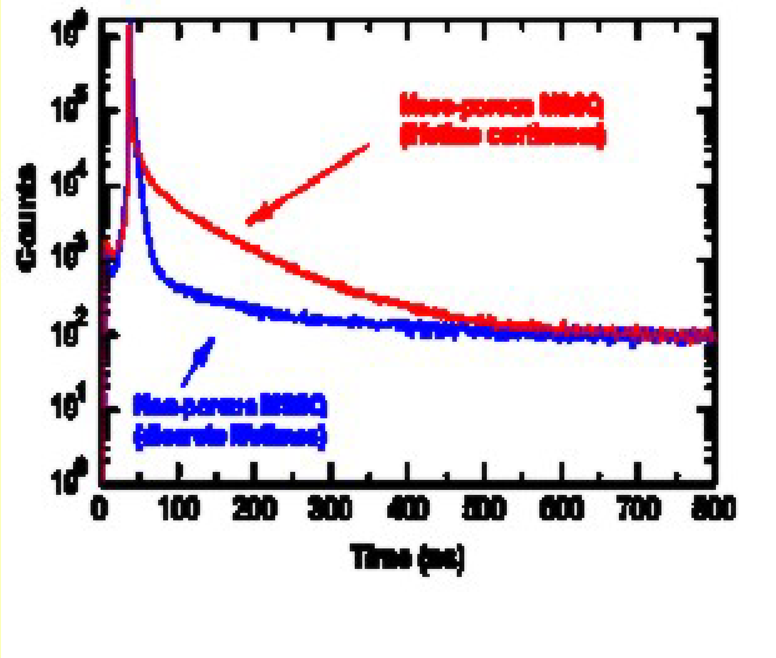
Thermalized Ps collides with the pore walls and the resulting Ps lifetime is shortened by positron annihilation with molecularly bound electrons in addition to the captured electron.

The reduced Ps lifetime is related to the pore size. A shorter lifetime will be produced if Ps is trapped in a smaller pore.

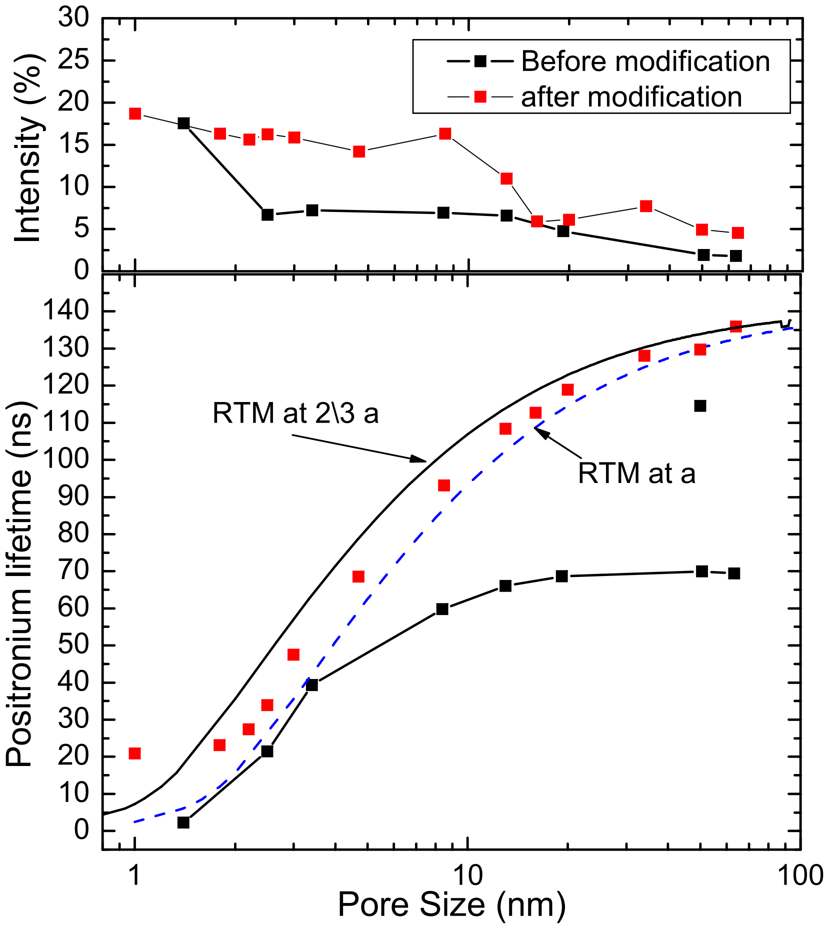
Furthermore, a distribution of Ps lifetimes may result if there is a distribution of pore sizes.

Only very short Ps lifetimes (1.5 and 6 ns) are fitted in the non-porous MSSQ (the nanovoids). The porous film, however, shows long-lived Ps events that can not be adequately fitted with a singer lifetime, i.e., a distribution of Ps lifetime is required.

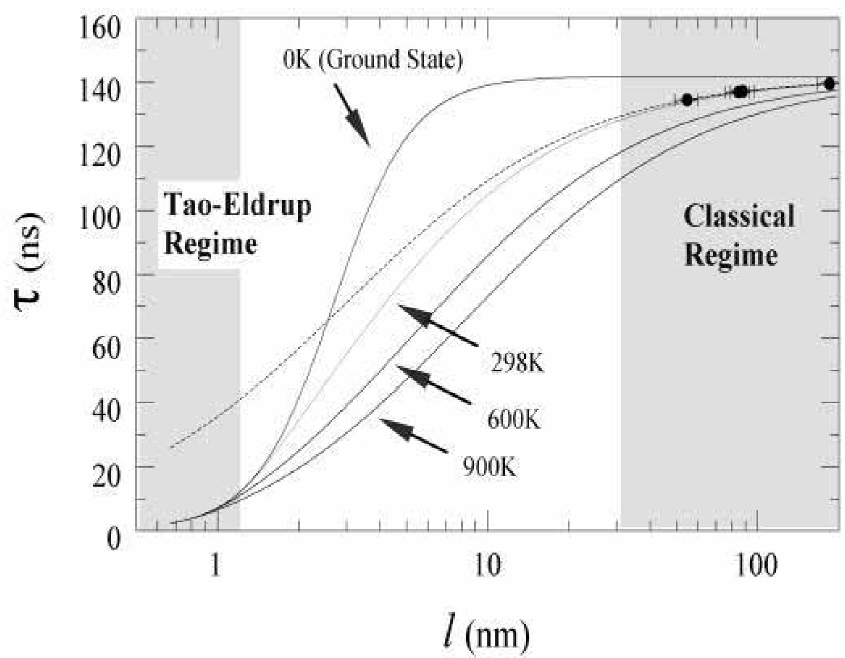




Calibration of Porous Solid By PALT

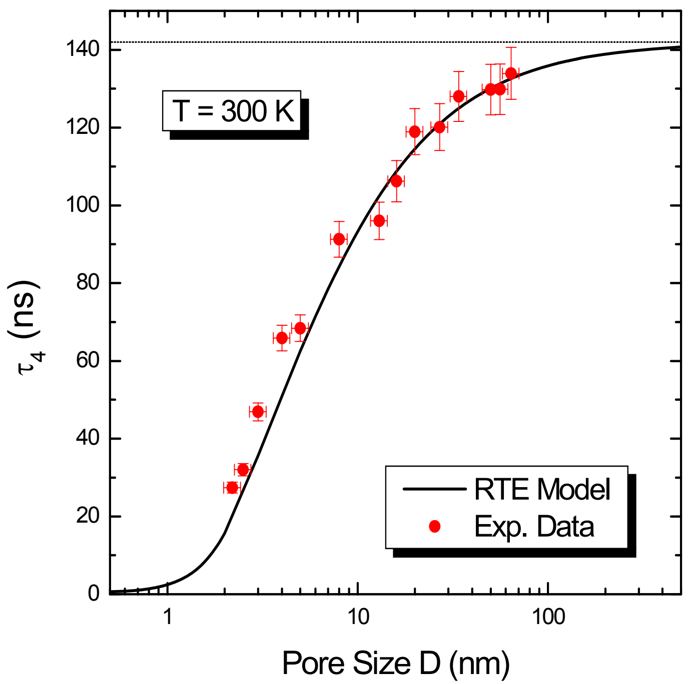


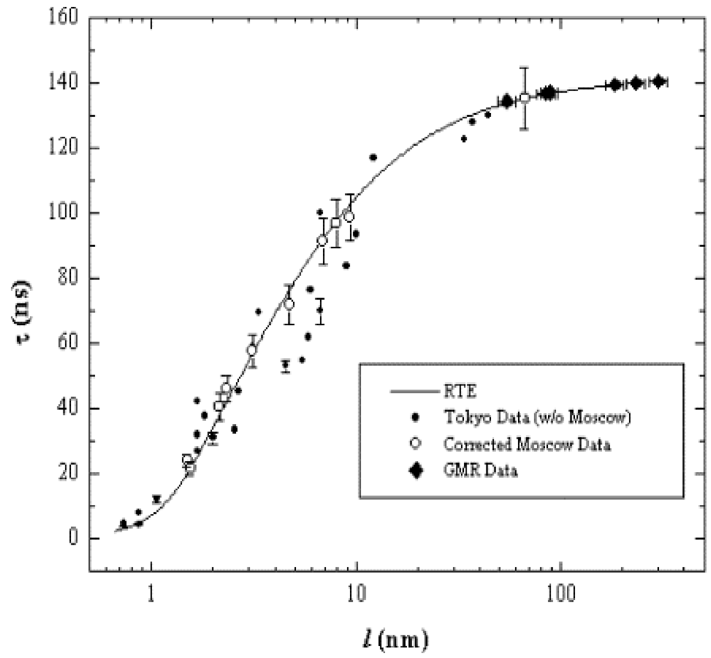
O-Ps lifetime t and the intensity I as a function of the pore size for porous glass. square (■) represent the experimental data at RT: the dash curve (-) is the theoretical results of RTE- model for the cylindrical geometry at **a** length and the solid curve for **2/3 a**.



T- dependence of the Ps Lifetime for cubical pores in the RTE model. The 0K (ground state) curve given by the Tao-Eldrup model and the dashed curve given by the classical model are presented for comparison . **Journal of Physical Chemistry B [Dull, 2001].**

Calibration of Porous Solid By PALT

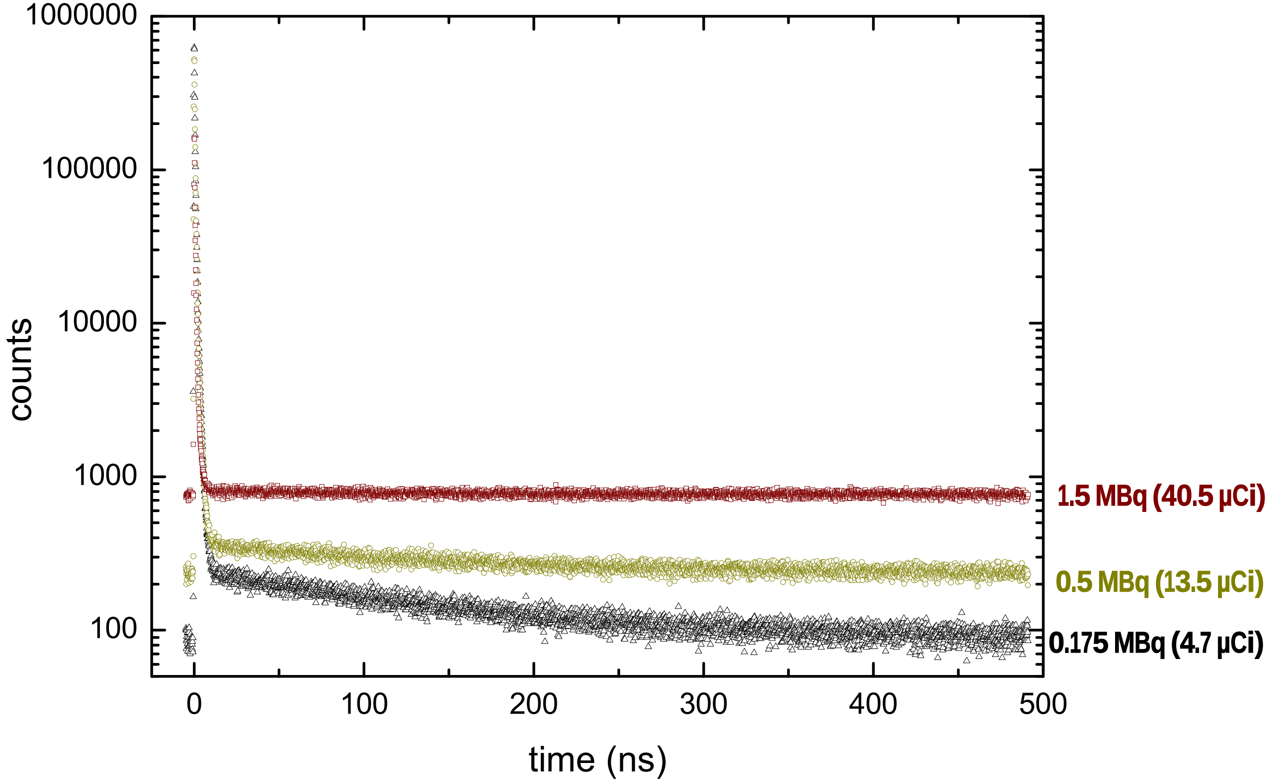




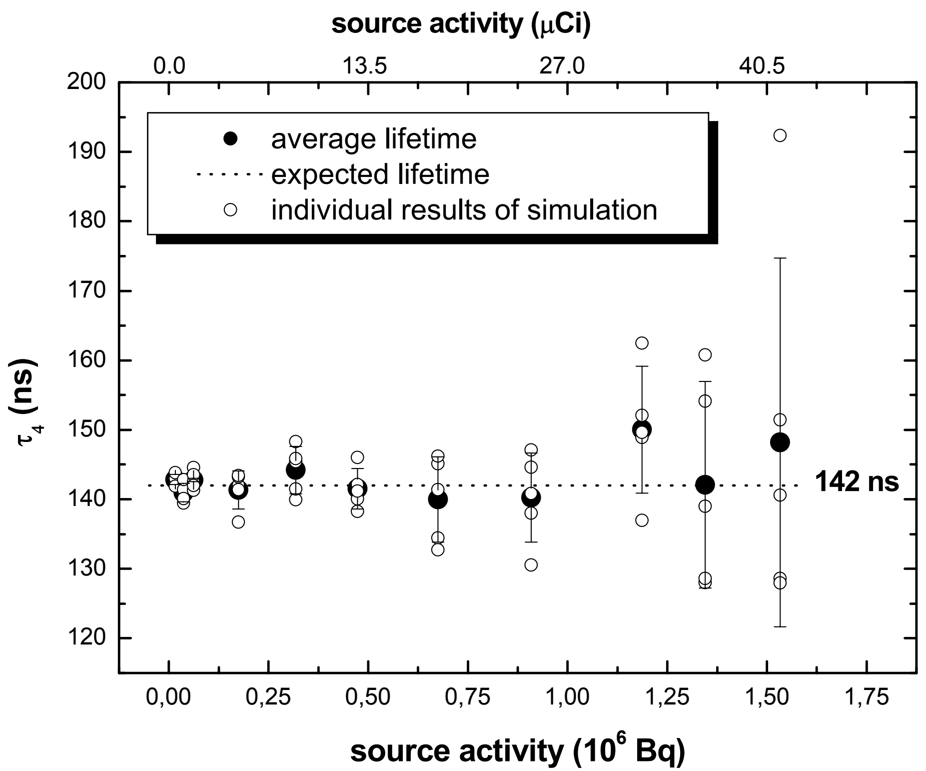
A comparison of the RTE model with experimental data acquired in bulk materials.

MCA -8000 channels
PMT -Hammarmatus
Plastic detectors
RSF≈ 220 ps

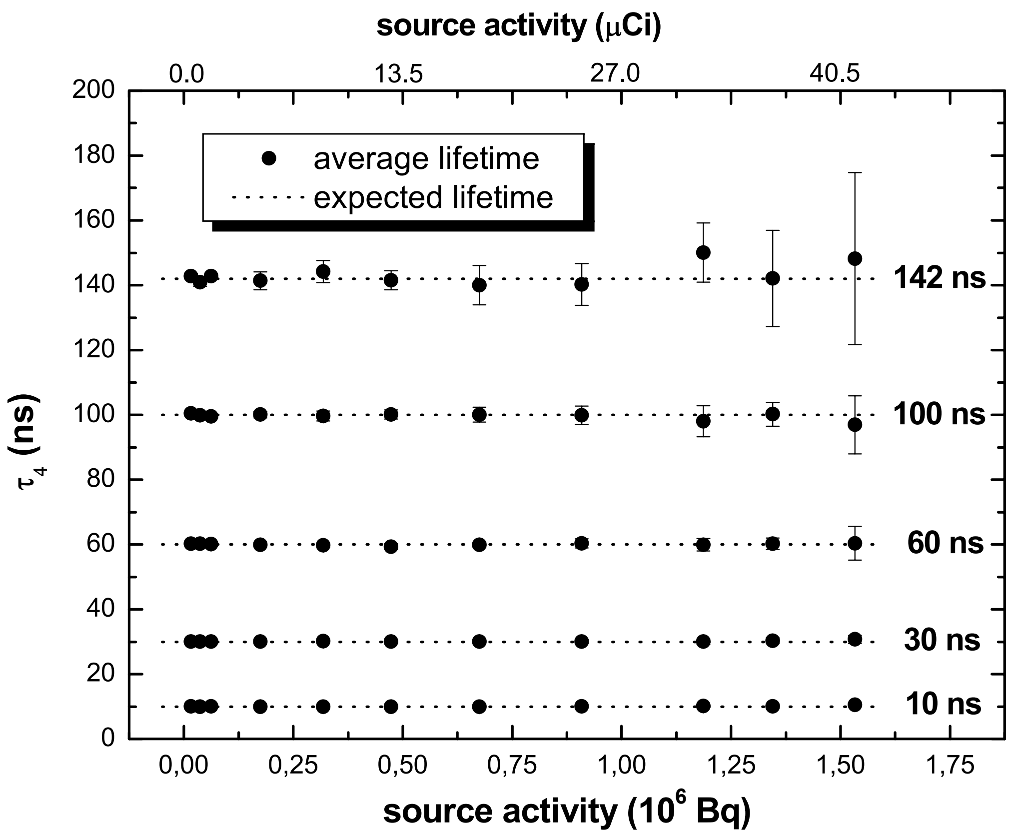
Simulation of the PALS spectra



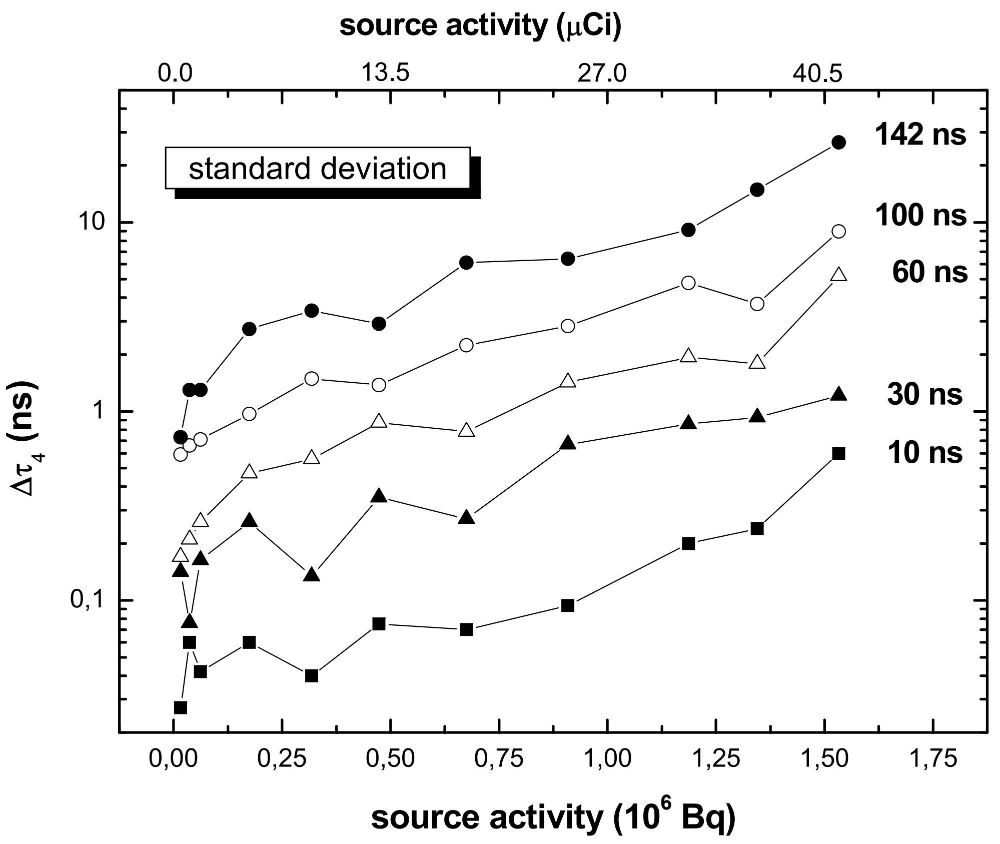
Simulation of the PALS spectra



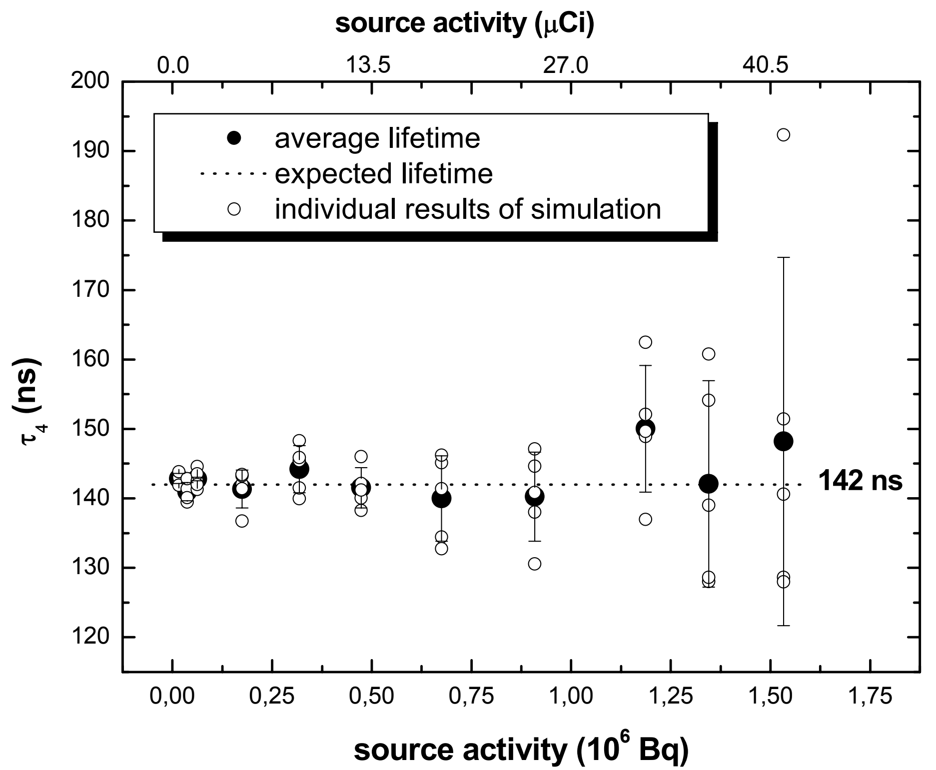
Simulation of the PALS spectra



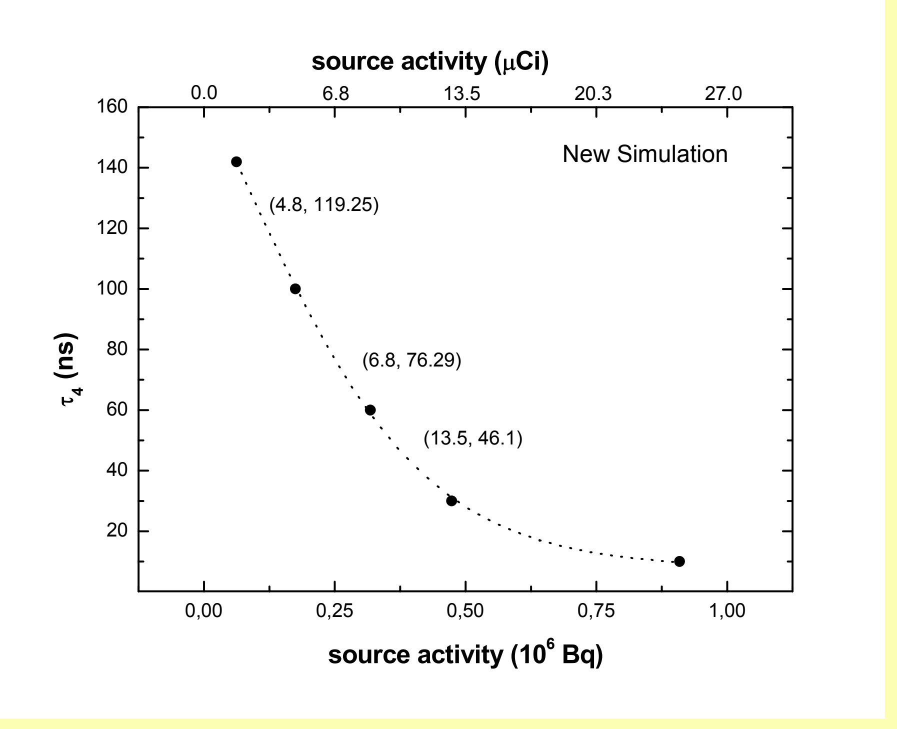
Simulation of the PALS spectra

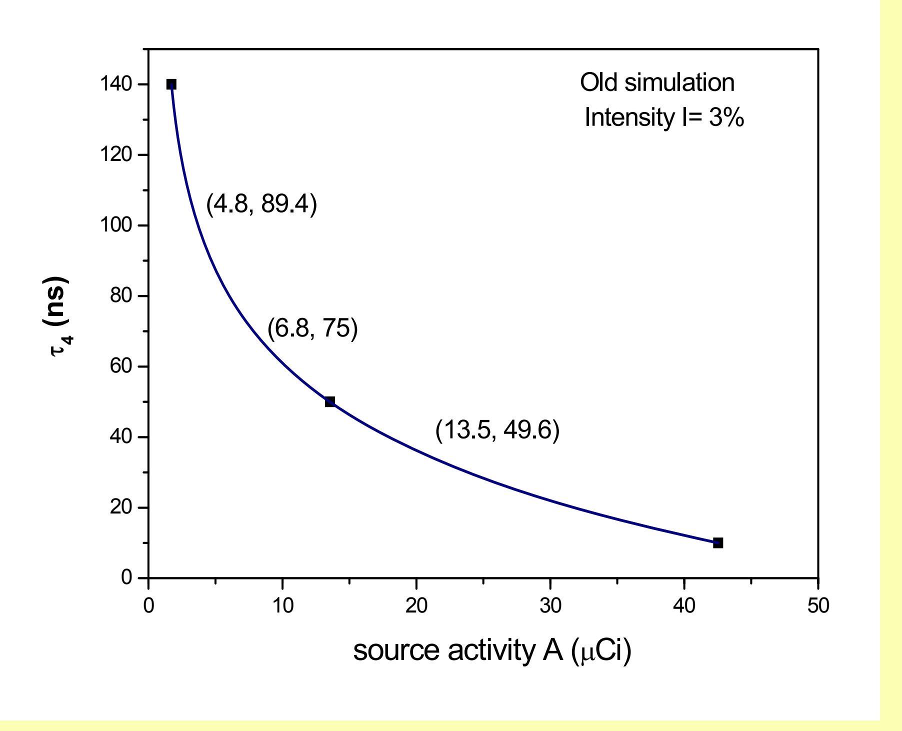


Simulation of the PALS spectra

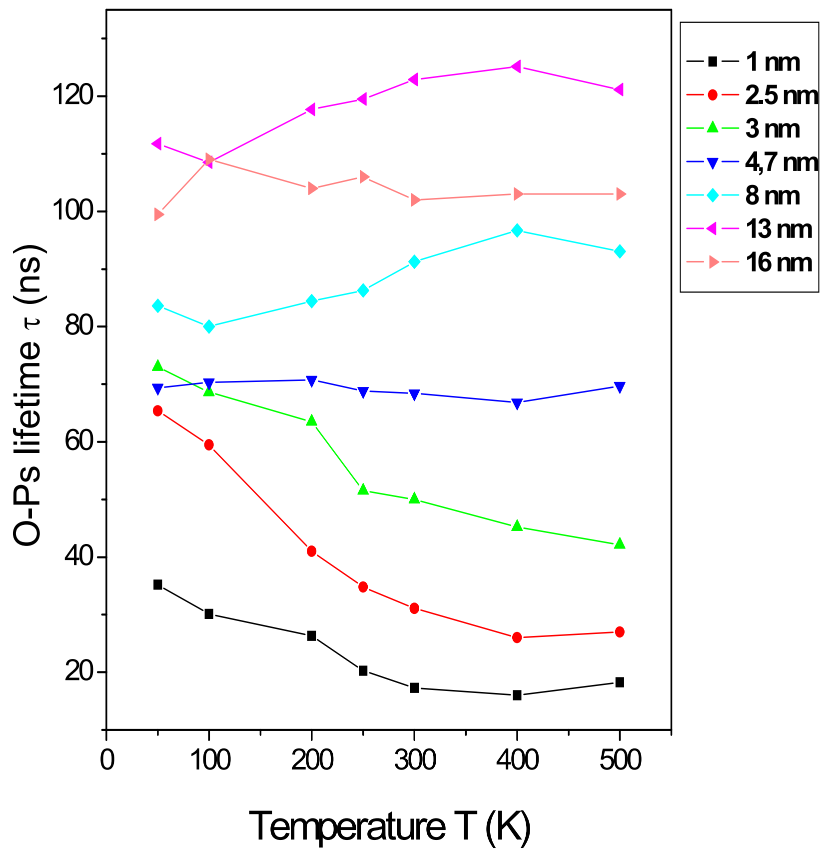


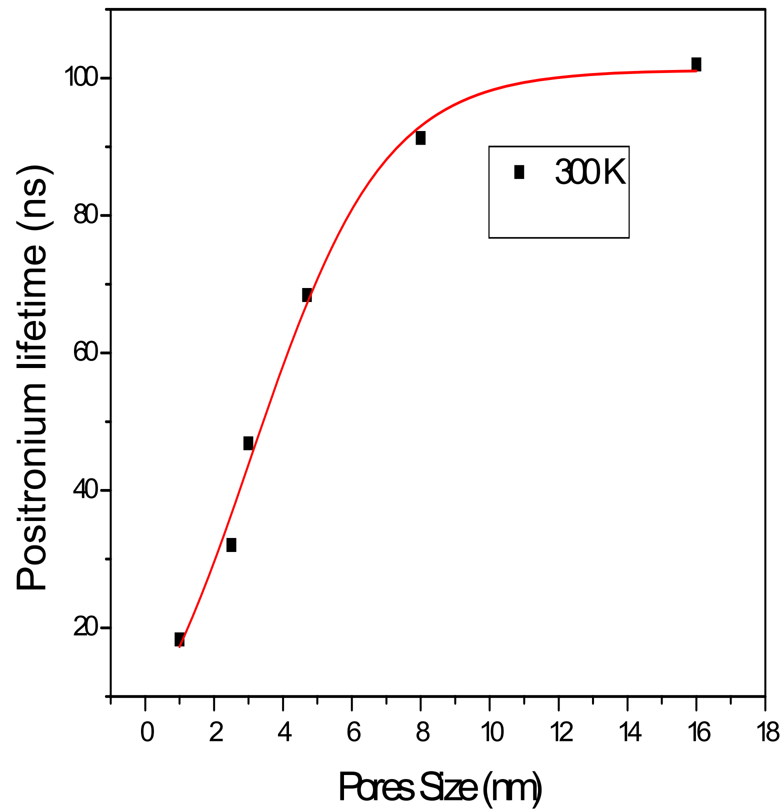
Simulation of the PALS spectra





T- Dependence Measurments





PHYSICAL REVIEW B VOLUME 60, NUMBER 8 15 A

Positronium annihilation in mesoporous thin films

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(Received 17 June 1999)

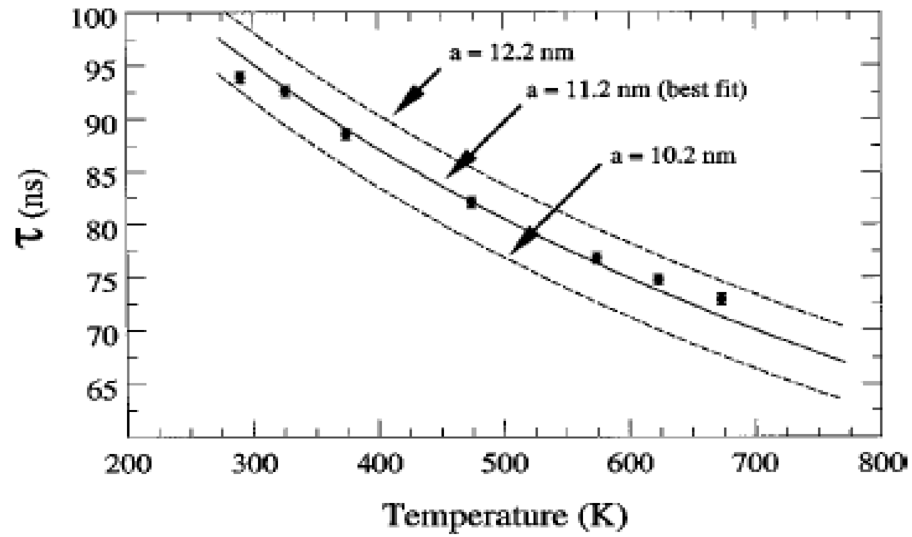

FIG. 3. The predicted temperature dependence for three different cubic pore sizes, *a*. The data shown are average values acquired during five heating cycles (to assure reproducibility and reversibility) of the TEOS-capped sol-gel film.

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Determination of Pore Size in Mesoporous Thin Films from the Annihilation Lifetime of Positronium

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Received: November 14, 2000; In Final Form: February 15, 2001

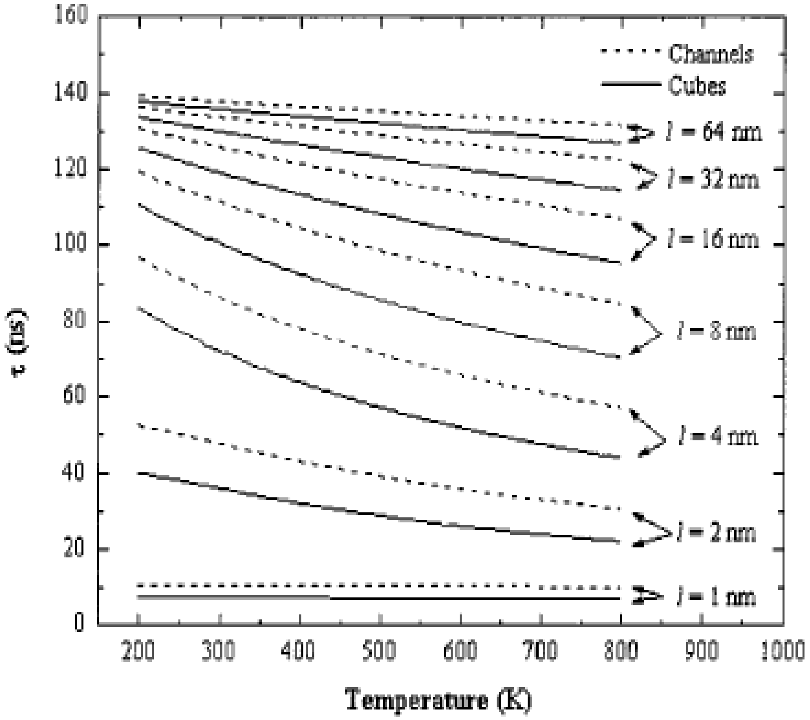


Figure 4. Ps lifetime vs temperature for a variety of mean free paths using both 2D and 3D pores in the calculation.

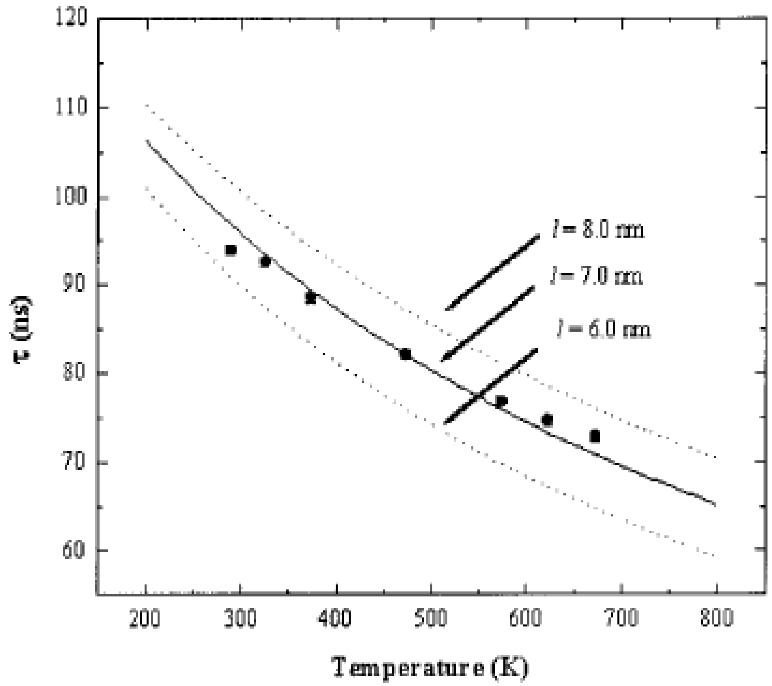


Figure 5. Ps lifetimes measured in a low-K porous silica film (Nanoglass A10B) as a function of film temperature and RTE curves (from ref 5).

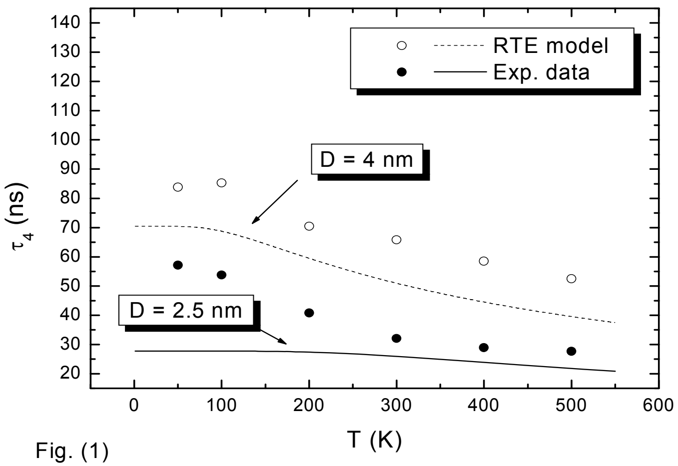


Fig. (1)

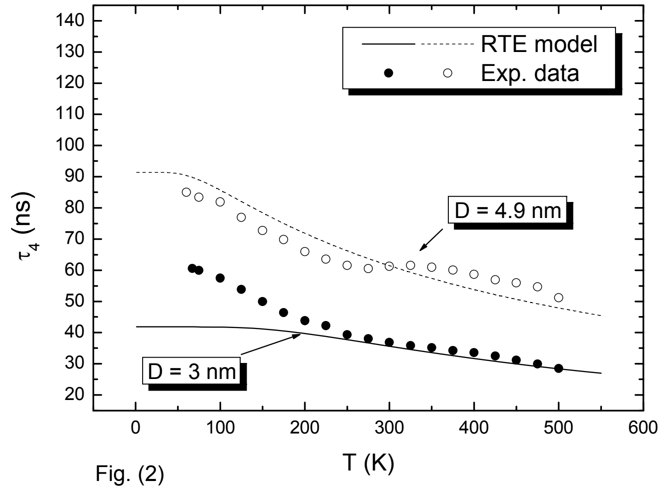


Fig. (2)

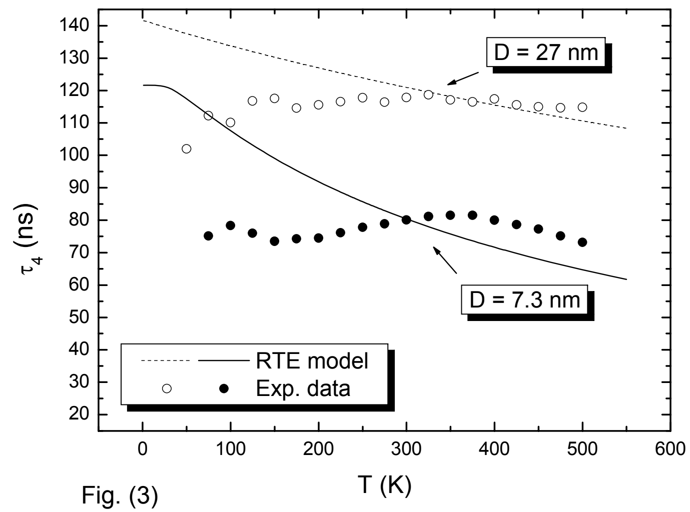
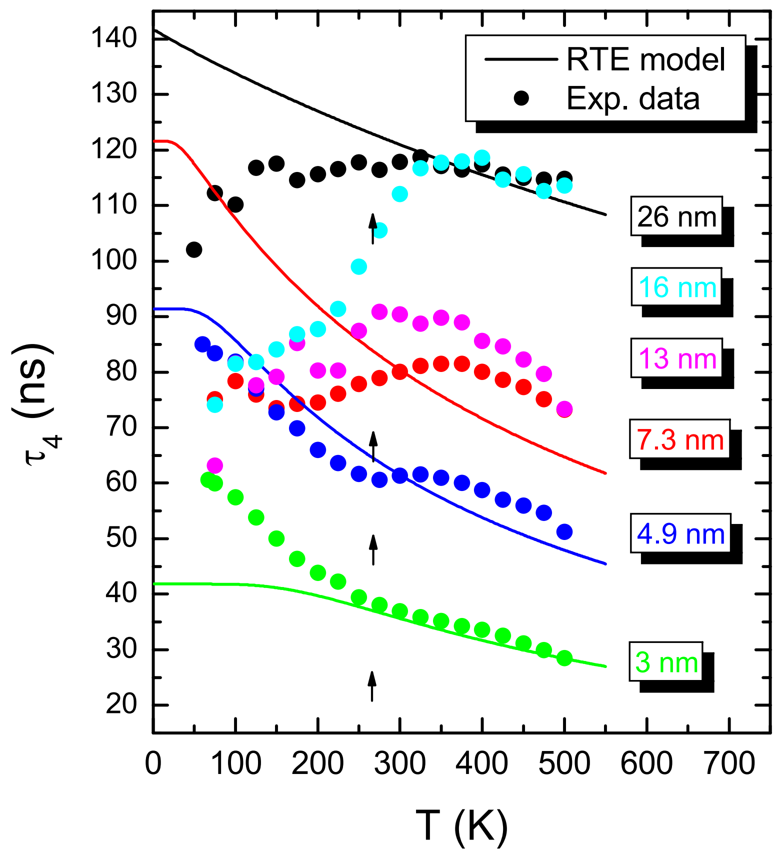
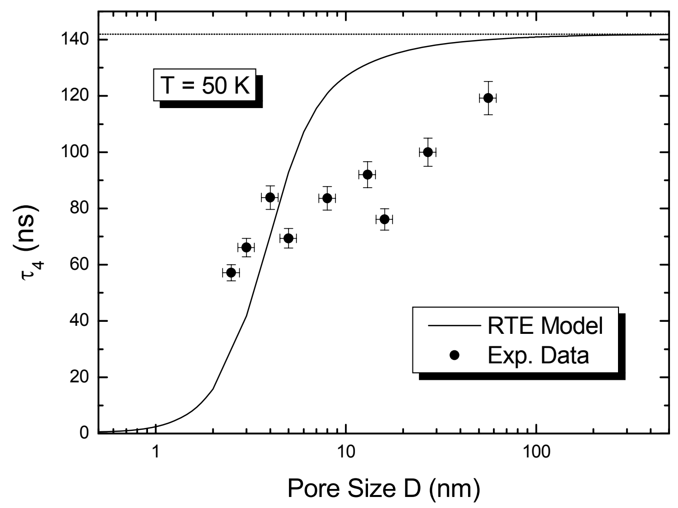
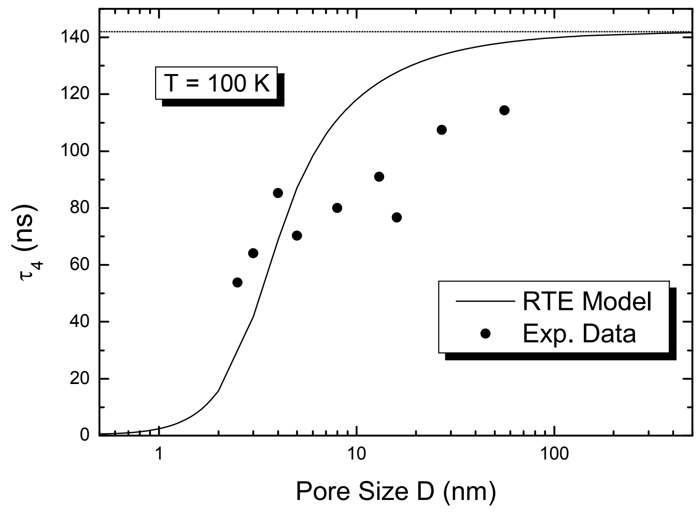


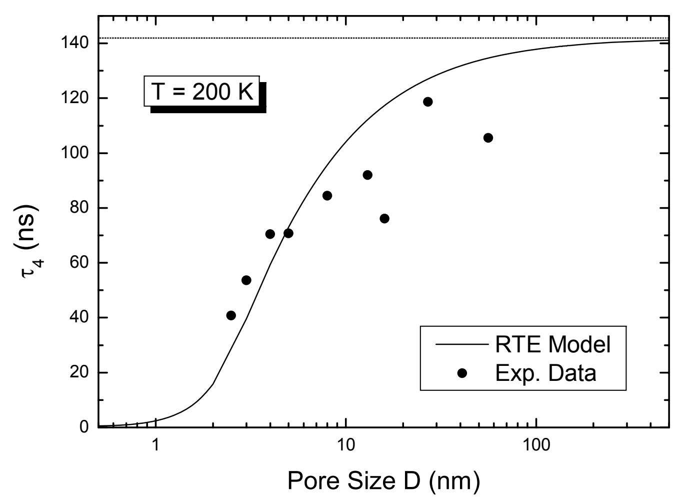
Fig. (3)

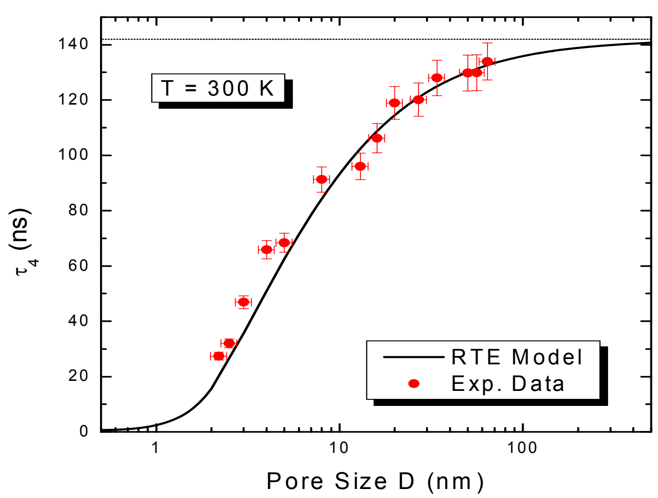


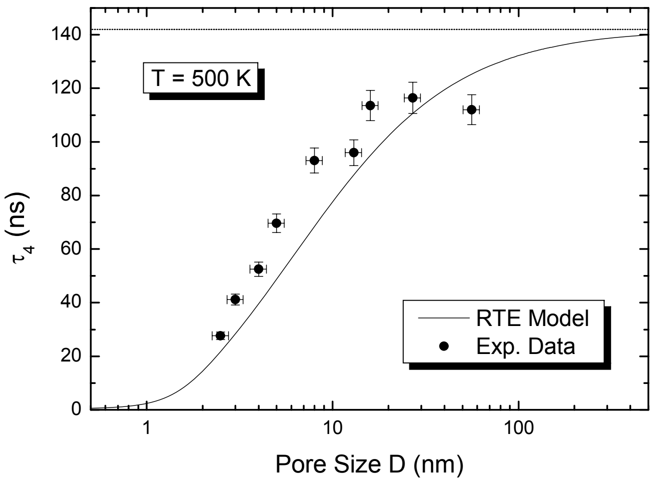
RTE model
Exp. data
26 nm
16 nm
13 nm
7.3 nm
4.9 nm
3 nm
τ_4 (ns)
T (K)

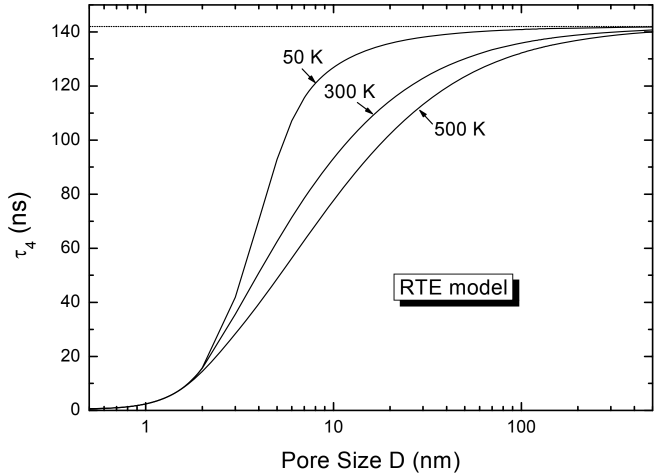

T = 50 K
RTE Model
Exp. Data
τ_4 (ns)
Pore Size D (nm)
0
20
40
60
80
100
120
140
1
10
100


T = 100 K
RTE Model
Exp. Data
τ_4 (ns)
Pore Size D (nm)
0
20
40
60
80
100
120
140
1
10
100


T = 200 K
RTE Model
Exp. Data
τ_4 (ns)
Pore Size D (nm)
0
20
40
60
80
100
120
140
1
10
100


T = 300 K
RTE Model
Exp. Data
τ_4 (ns)
Pore Size D (nm)
0
20
40
60
80
100
120
140
1
10
100





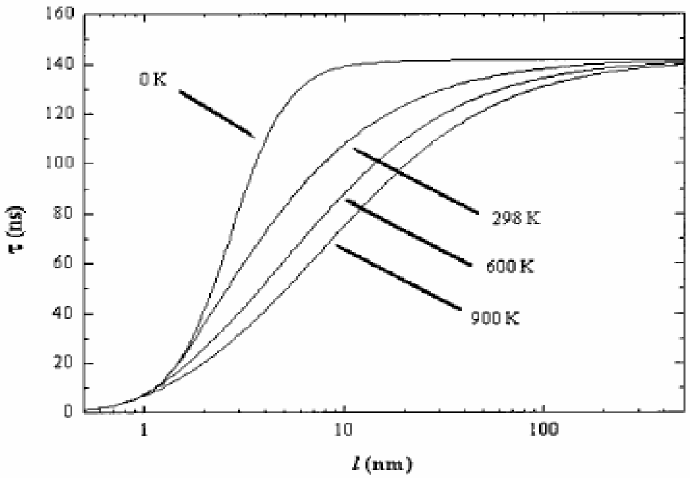
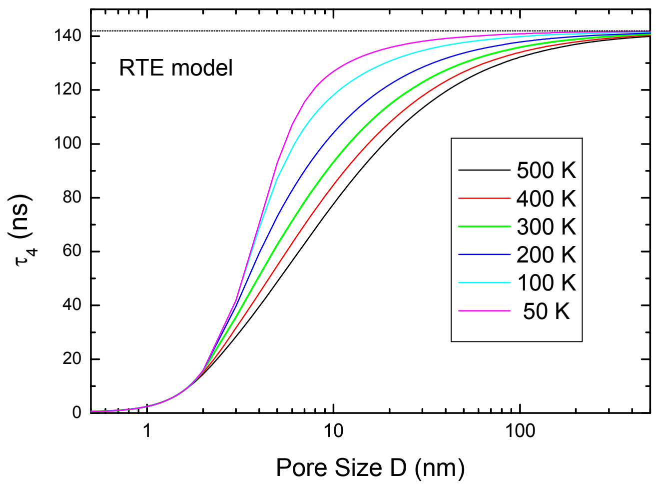


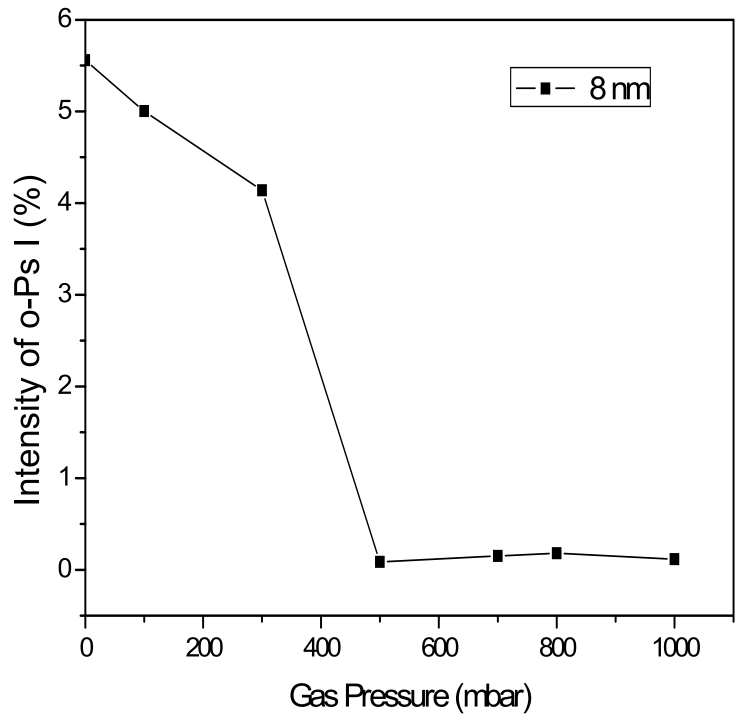
Figure 3. Temperature dependence of the Ps lifetime for cubical pores in the RTE calculation. The mean free path, l, is related to the cube side length by $l = {}^2/_3 a$.



Cryo-Condensation Effect

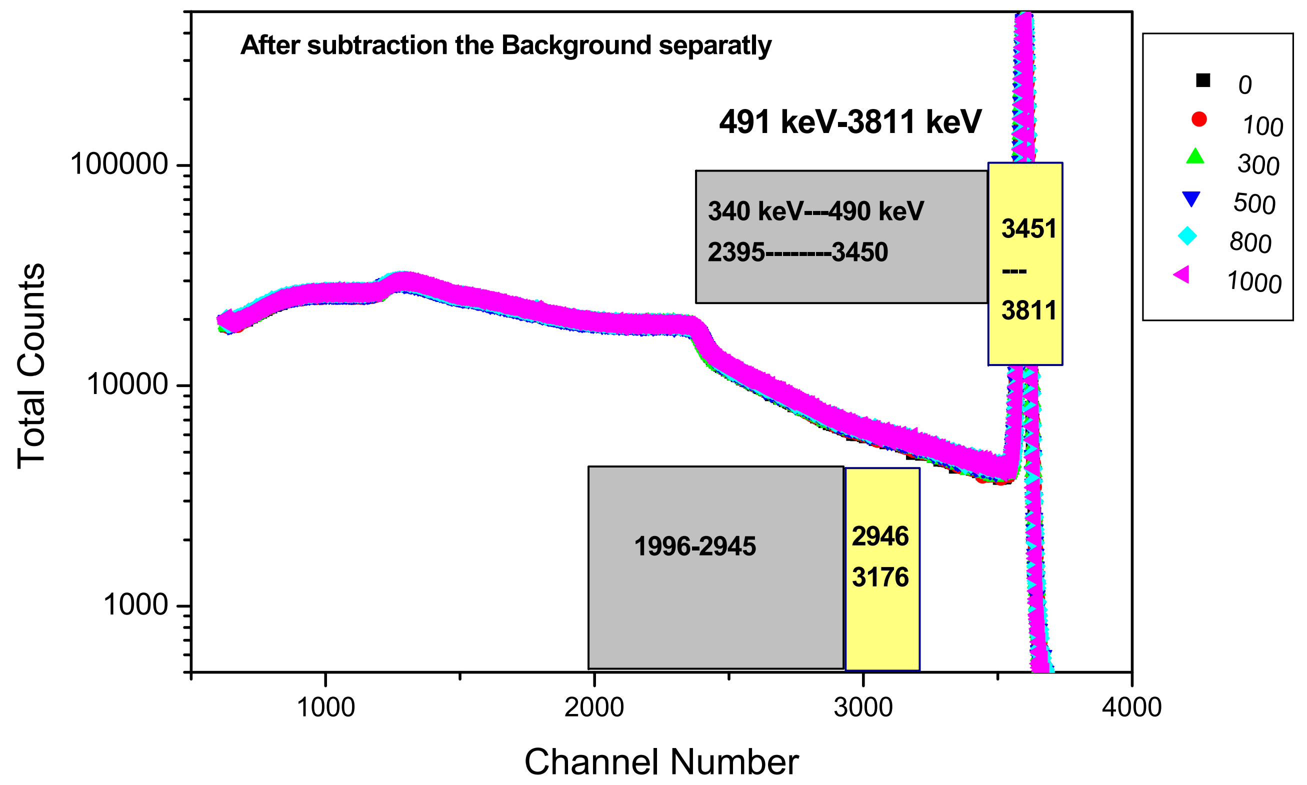
- **Lifetime Measurments:**

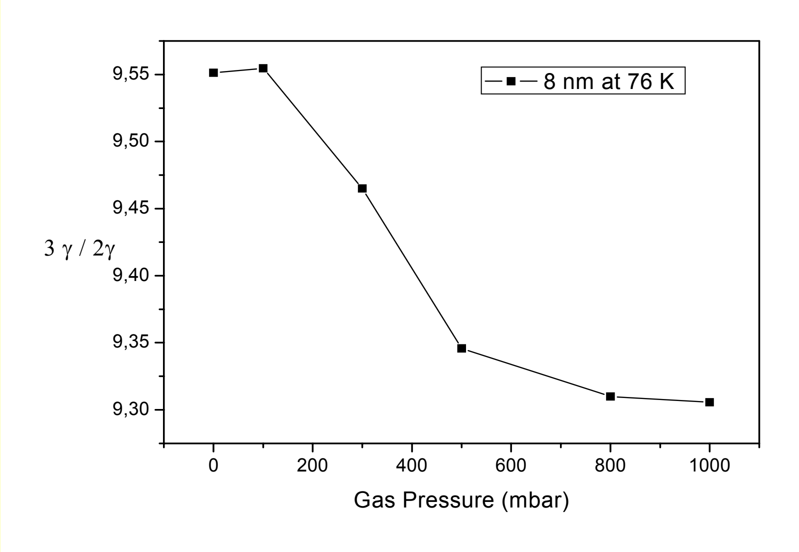
P (mbar)	t_4	I_4
0	97.1	5.99
100	109.1	5.003
300	124.27	4.141
500	236.6	0.084
700	79.9	0.149
800	150.4	0.179
1000	0.163	0.116



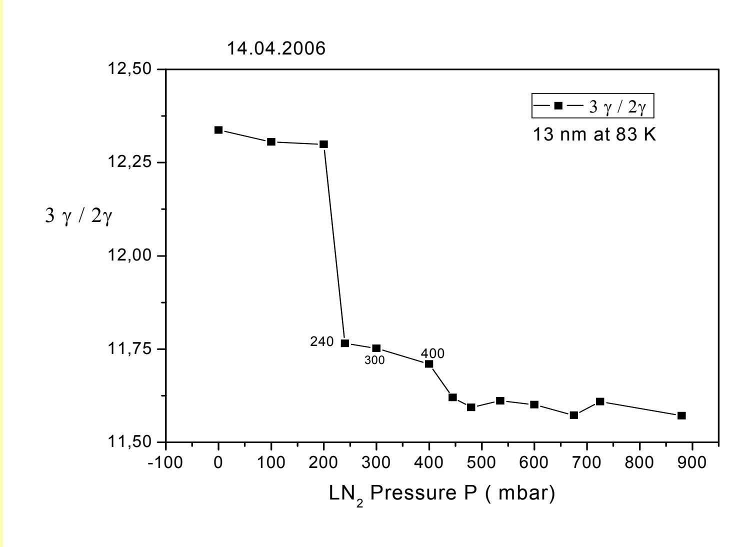
4000 channels
XP2020
4 components

Doppler Broadening Measurments

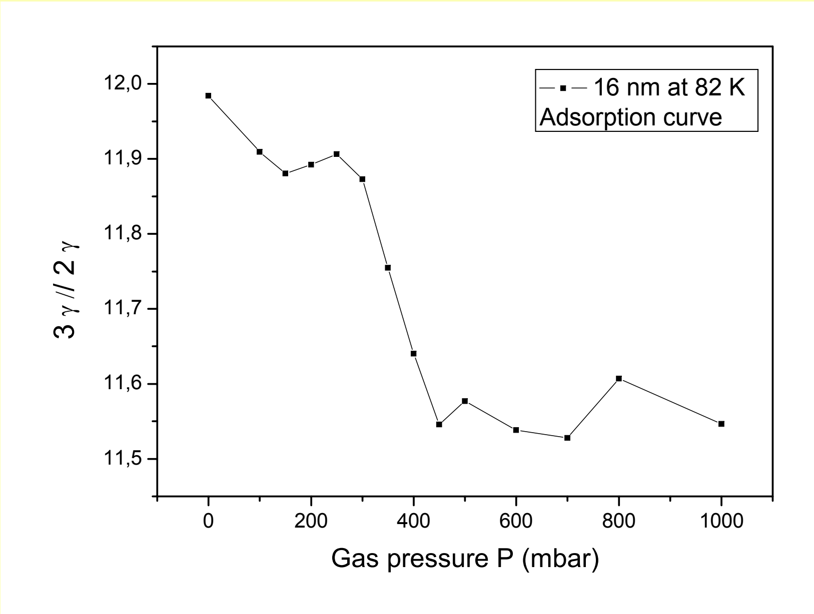




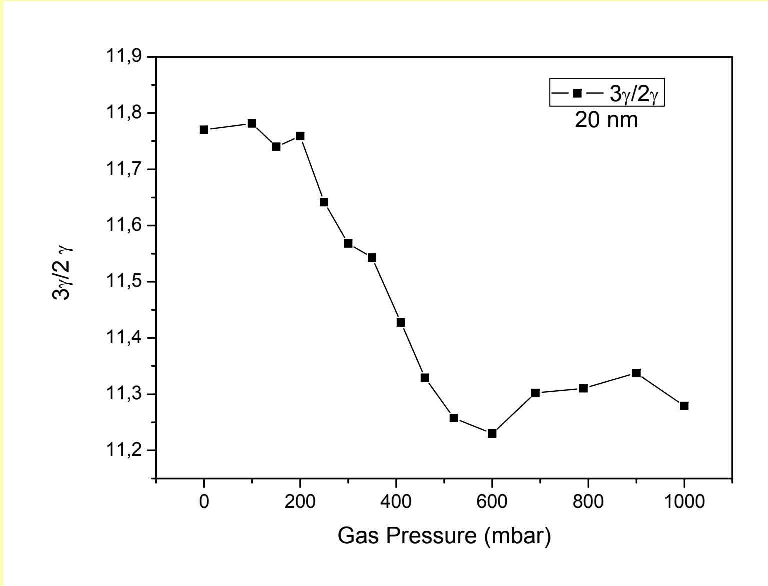
8 nm at 76 K
3 γ / 2γ
9,55
9,50
9,45
9,40
9,35
9,30
0
200
400
600
800
1000
Gas Pressure (mbar)



14.04.2006
3 γ / 2γ
13 nm at 83 K
3 γ / 2γ
12,50
12,25
12,00
11,75
11,50
240
300
400
-100
0
100
200
300
400
500
600
700
800
900
LN2 Pressure P (mbar)



16 nm at 82 K
Adsorption curve
3 γ / 2 γ
12,0
11,9
11,8
11,7
11,6
11,5
0
200
400
600
800
1000
Gas pressure P (mbar)



3γ/2γ
20 nm
3γ/2 γ
11,9
11,8
11,7
11,6
11,5
11,4
11,3
11,2
0
200
400
600
800
1000
Gas Pressure (mbar)

Thanks for you attention

www.Positron.physik.uni-halle.de