Verifying the RTE model – ortho-positronium lifetime measurement on controlled pore glasses

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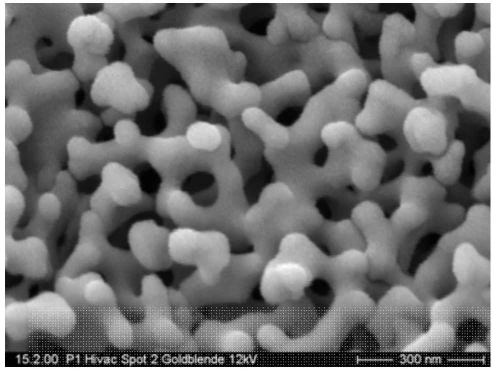
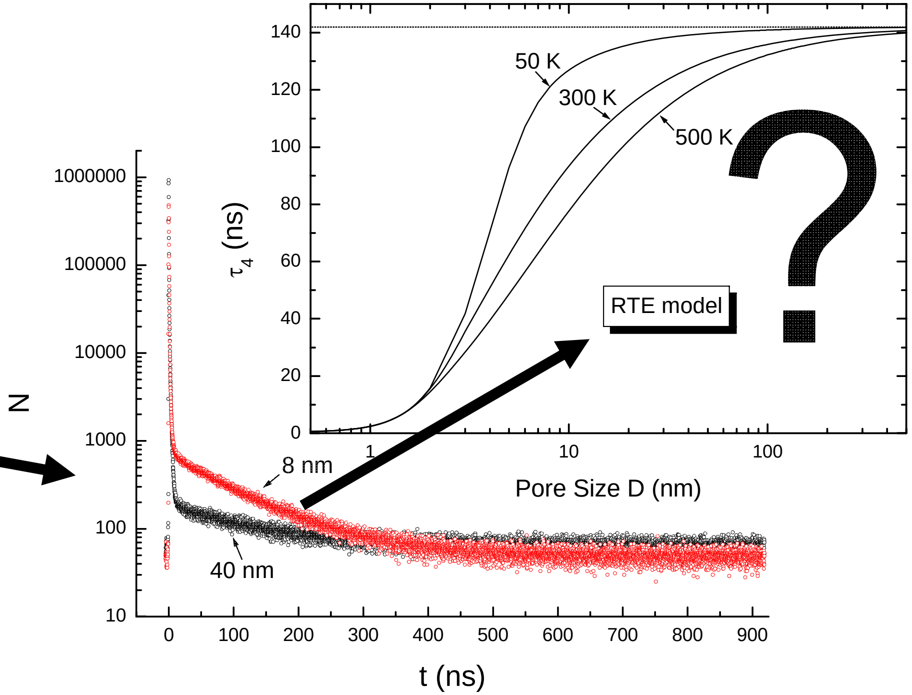
Verifying the RTE model

- Porous glass



- Models – the state of the art
- Experimental results



- Summary

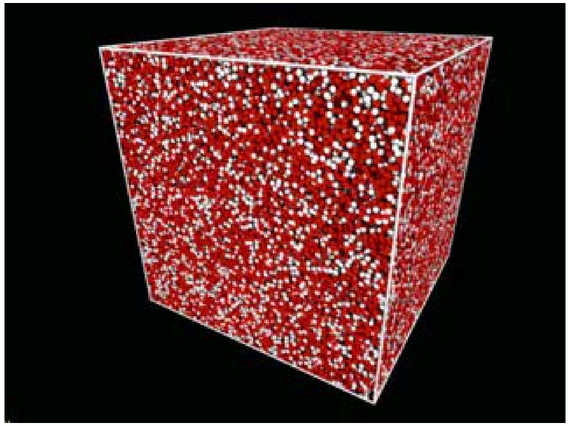
Controlled pore glasses - CPG

VYCOR-Process

alkali borosilicate glass

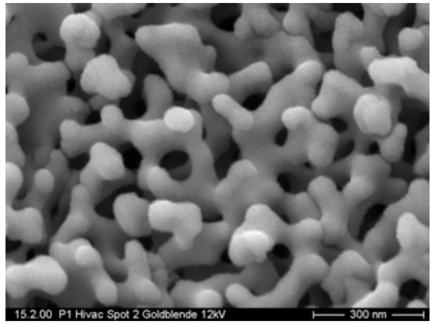
T

530 - 710°C



Extraction

HCl/NaOH

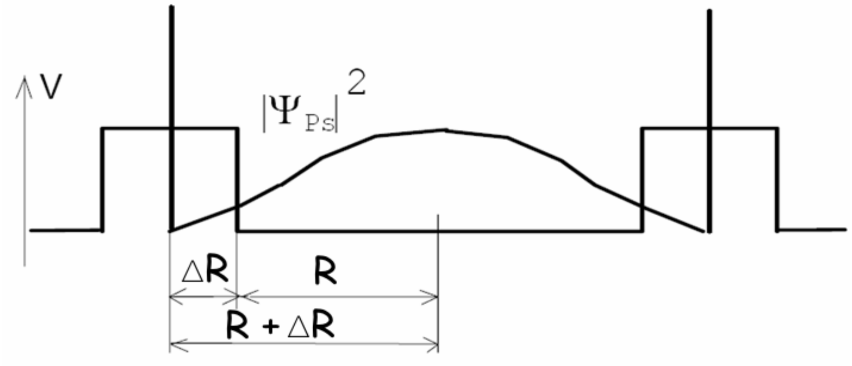


d_p 1 to 110 nm

- spinodal phase separation
- decomposition is initiated by heat treatment
- alkali rich borate phase <-> pure silica
- alkali phase soluable in acid -> silica network
- pore size depends on basic material
- shape depends on duration and T of heat treatment

F. Janowski, D. Enke in F. Schüth, K.S.W. Sing, J. Weitkamp (Eds.), *Handbook of Porous Solids*, WILEY-VCH, Weinheim, **2002**, 1432-1542.

The TE model



- **Annihilation rate:** $\frac{1}{\tau_{o-Ps}} = \lambda_{o-Ps}$

$$= \lambda_{2\gamma} + \lambda_{3\gamma}$$

$$= \lambda^0_{2\gamma}(P) + \lambda^0_{3\gamma}(1-P) \cong \lambda^0_{2\gamma}(P)$$

$$\lambda^0_{2\gamma} = \frac{\lambda_S + 3\lambda_T}{4} = \lambda_A \approx 2ns^{-1}$$

- **Pore size < 1 nm -> $\lambda_{3\gamma}$ neglected, only pick off annihilation**

$$\lambda_{TE}(R) = \lambda_A\left[1 - \frac{R}{R+\Delta R} + \frac{1}{2\pi}\sin\left(\frac{2\pi R}{R+\Delta R}\right)\right]$$

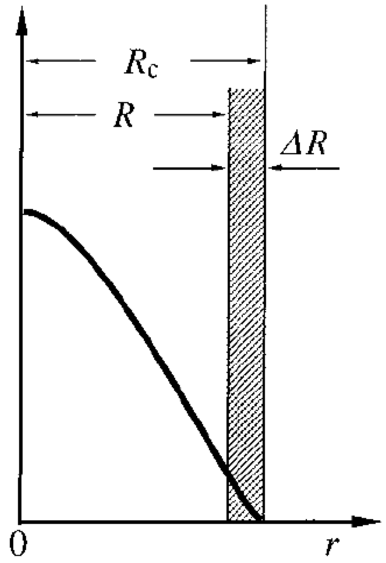
- ΔR **= 0.166 nm determined by Eldrup**
- **Pore size > 1 nm -> $\lambda_{3\gamma}$ can not be neglected, temperature dependence of o-Ps lifetime (excited states)**

Tao, S. J. *J. Chem. Phys.* **1972**, *56*, 5499-5510. / Eldrup, M.; Lightbody, D.; Sherwood, J. N. *Chem. Phys.* **1981**, *63*, 51-58.

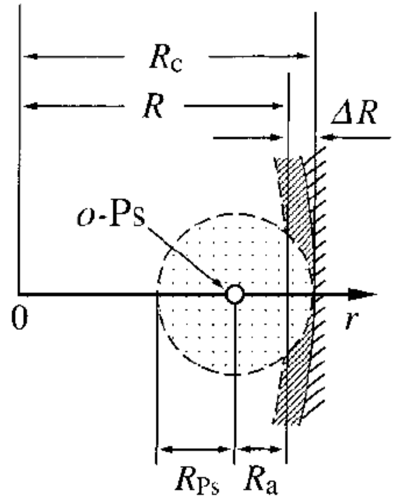
The 2 models for R > 1 nm - Tokyo

- **Tokyo model:** $$\lambda_{Tokyo}(R) = \begin{cases} \lambda_{TE} + \lambda_{3\gamma} & (R < R_a) \\ \lambda_{TE}(R_a)\left[1 - \left(\frac{R - R_a}{R + \Delta R}\right)^b\right] + \lambda_{3\gamma} & (R \geq R_a) \end{cases}$$

  - <u>Problems:</u> - no explicit temperature dependence
    - two free parameters to be determined



(a) TE



(b) Tokyo

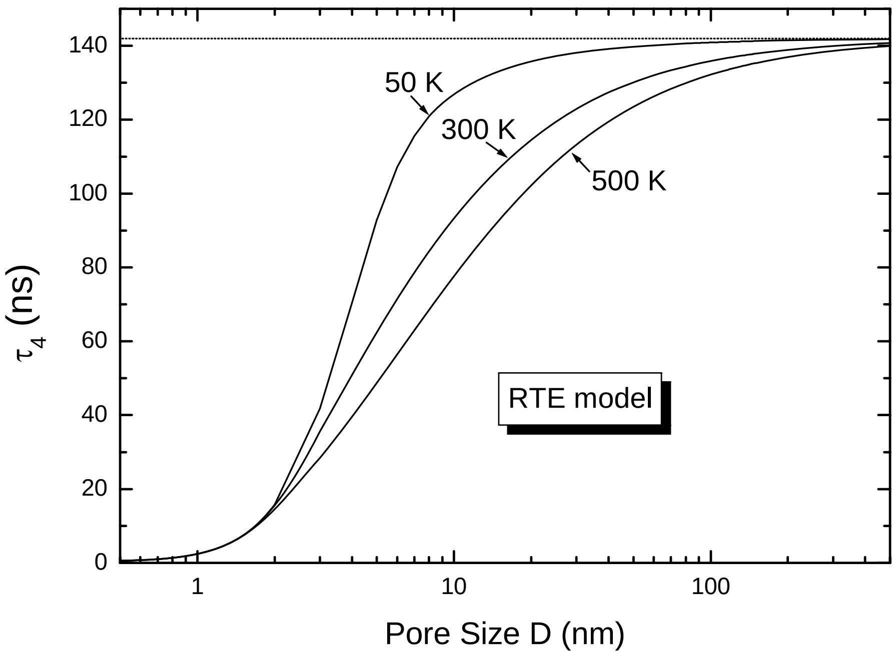
empirical:
R_a = 0.8 nm
b = 0.55

K. Ito, H. Nakanishi, Y. Ujihira, *J. Phys Chem. B* **1999**, *103,* 4555.

The 2 models for R > 1 nm - RTE

- **RTE model (for 3D cubic pores):**

$$\lambda_{RTE}(D,T) = \lambda_A - \frac{\lambda_S - \lambda_{3\gamma}}{4}\left[1 - \frac{2\delta}{D} + \frac{\sum_{i=1}^{\infty} \frac{1}{i\pi} \sin\left(\frac{2i\pi\delta}{D}\right) e^{\left(\frac{-\beta i^2}{D^2 kT}\right)}}{\sum_{i=1}^{\infty} e^{\left(\frac{-\beta i^2}{D^2 kT}\right)}}\right]^3$$



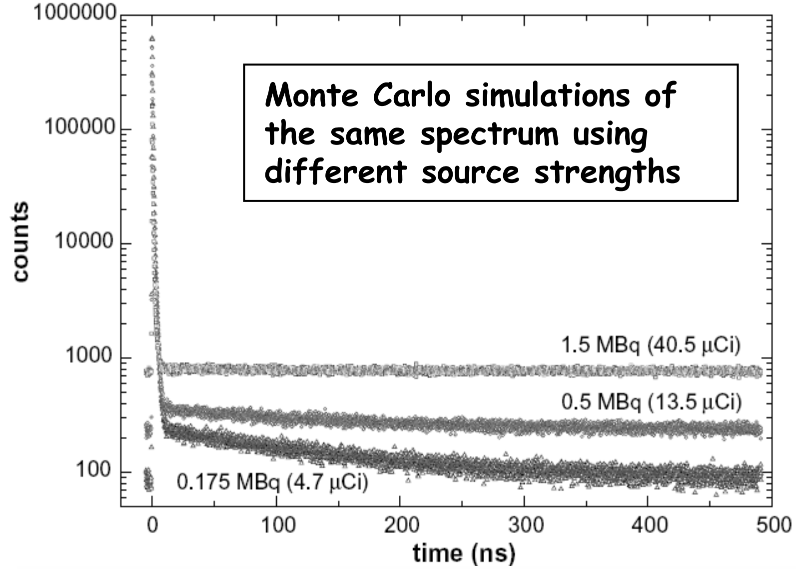
- **Boltzmann statistics ascribes explicit temperature dependence to the lifetime**
- **Rectangular geometry -> prevention of complicated Bessel functions**
- **δ = 0.18 nm analogous to TE model**

D. W. Gidley, T. L. Dull, W. E. Frieze, J. N. Sun, A. F. Yee, *J. Phys. Chem. B* **2001**, *105*, 4657.

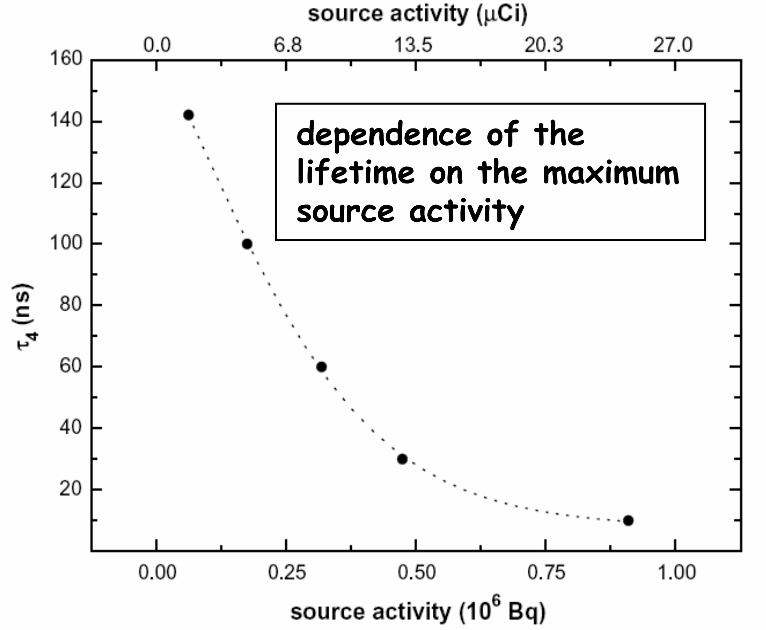
Verifying the RTE model

The experiments

- **Important: weak source required to obtain o-Ps lifetime properly (long lifetime component disturbed by chance coincidences)**



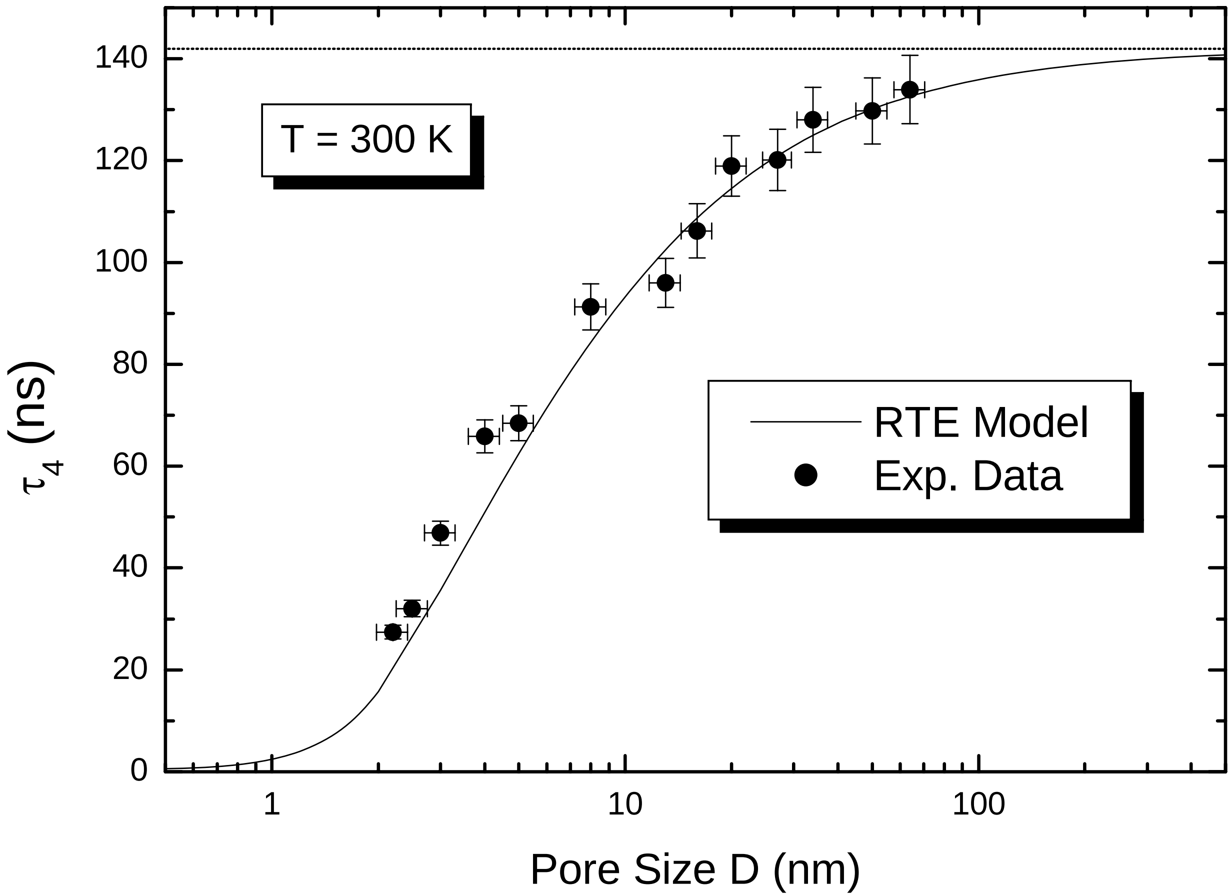
- **When expecting a lifetime of e.g. 120 ns -> max. source strength of 3 μCi recommended**



- **At first measurements at T = 300 K on different pore sizes**

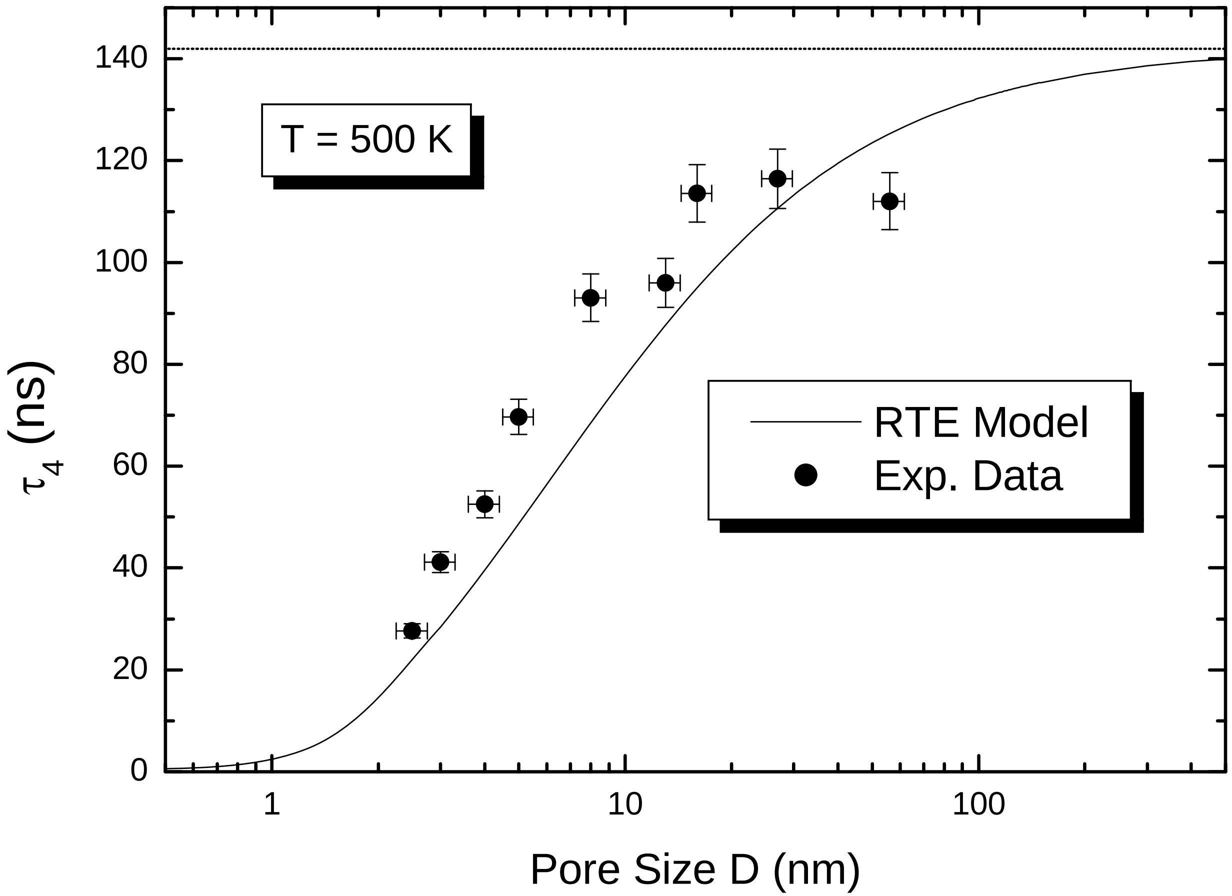
S. Thraenert, E.M. Hassan, R. Krause-Rehberg, *Nucl. Instrum. and Meth. B* **2006**, *Vol. 248 No. 2,* 336.

The experiments at T = 300 K



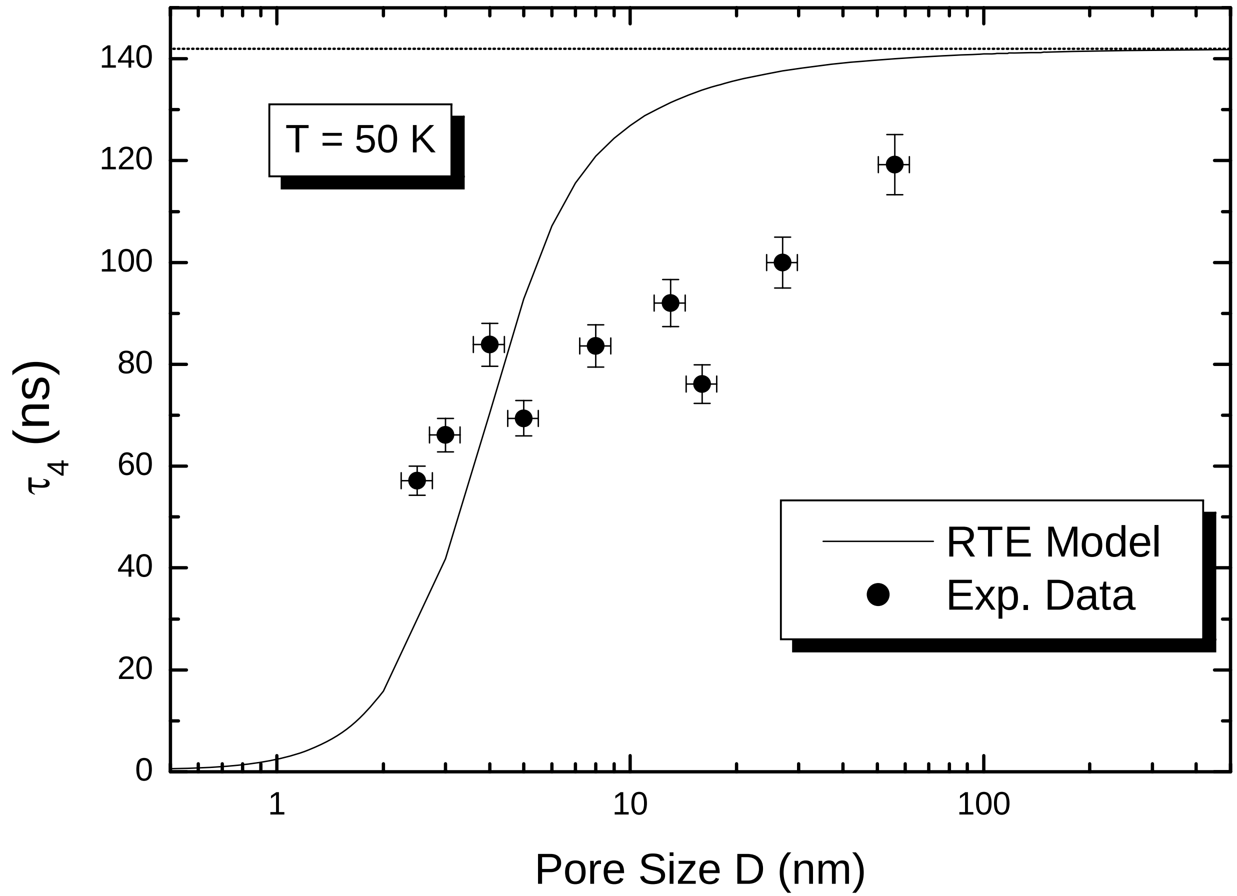
- **for T = 300 K general agreement to the RTE model**
- **calibration curve for the correlation of o-Ps lifetime and pore size**

The experiments at T = 500 K



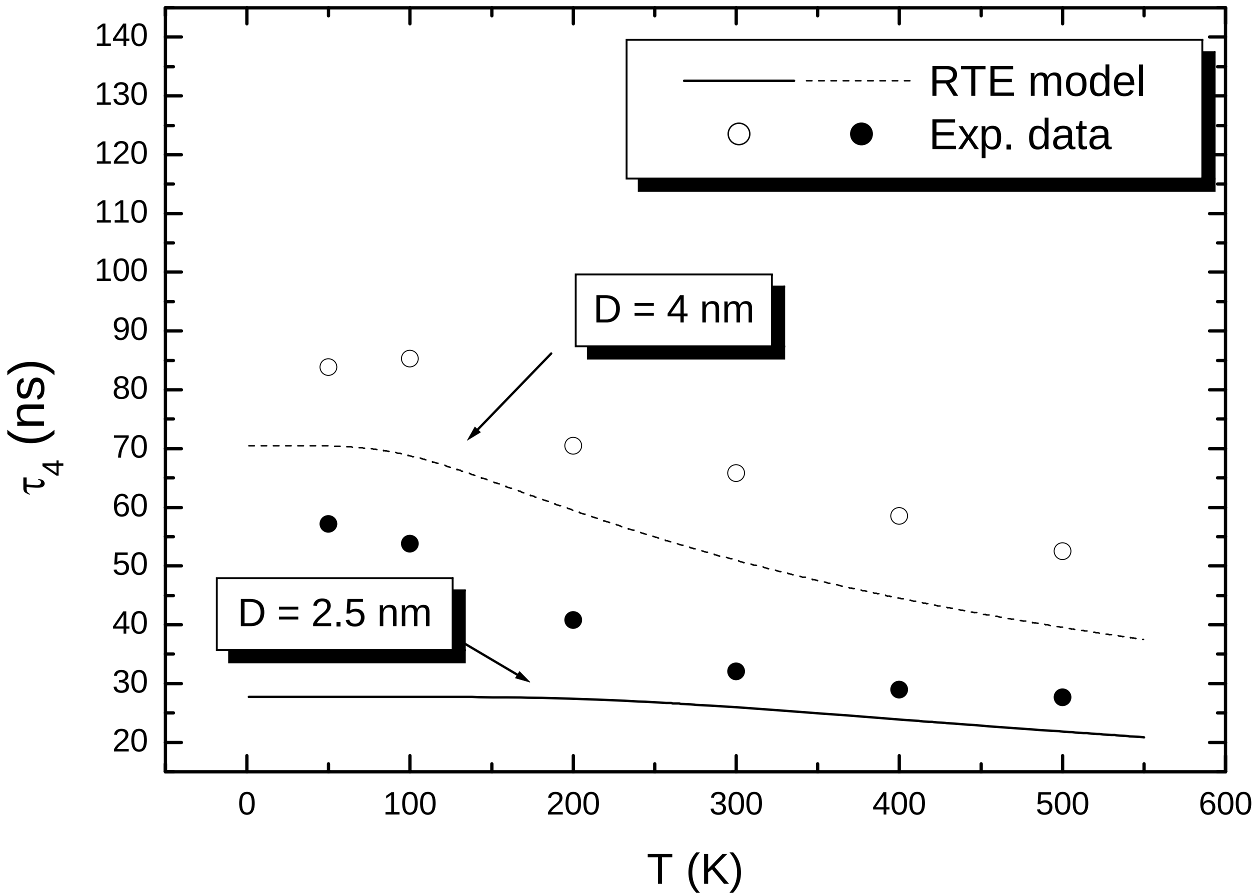
- **for T = 500 K still acceptable agreement to the RTE model**

The experiments at T = 50 K



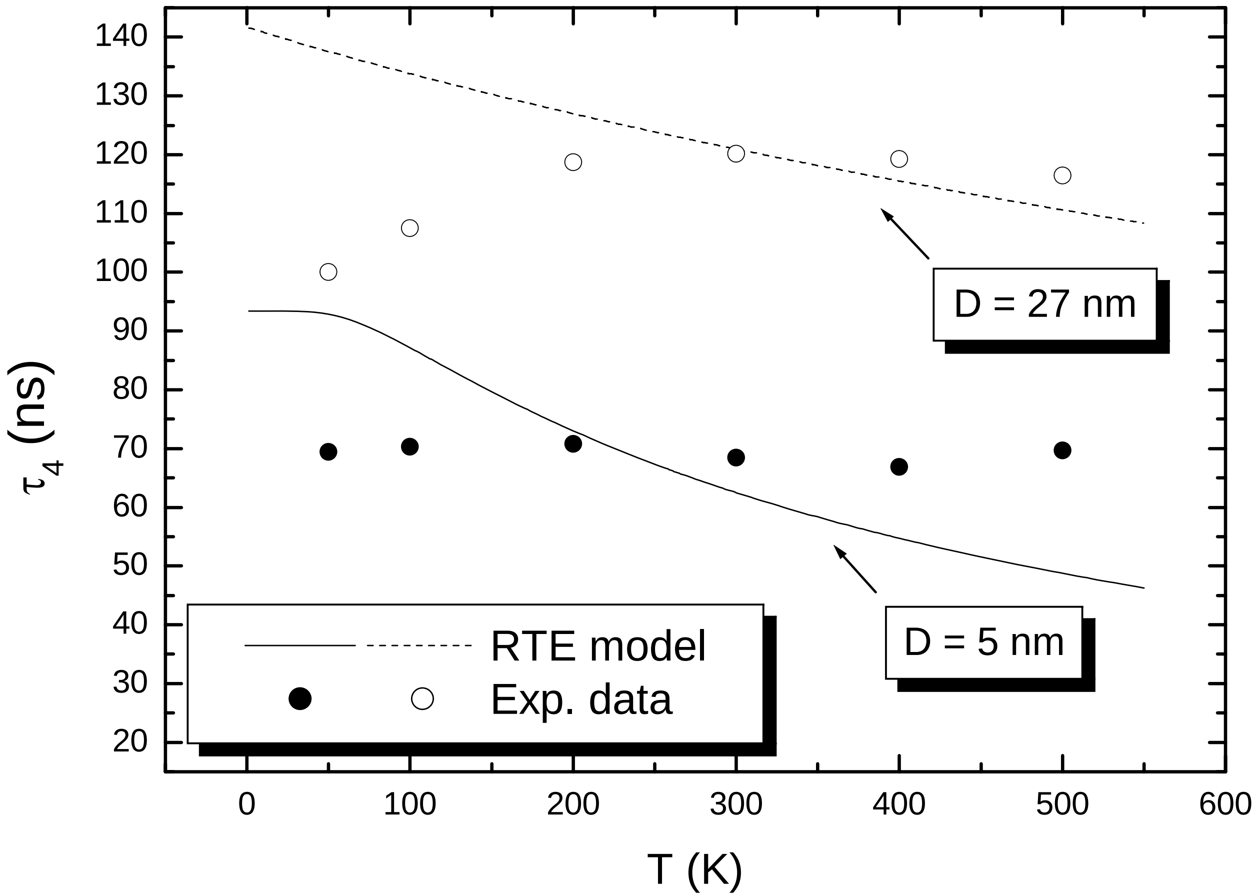
- **for T = 50 K no agreement to the RTE model can be observed**

The temperature dependence



- for D = 2.5 nm poor agreement to RTE model for T < 300 K
- for D = 4 nm shape of exp. data like RTE model

The temperature dependence



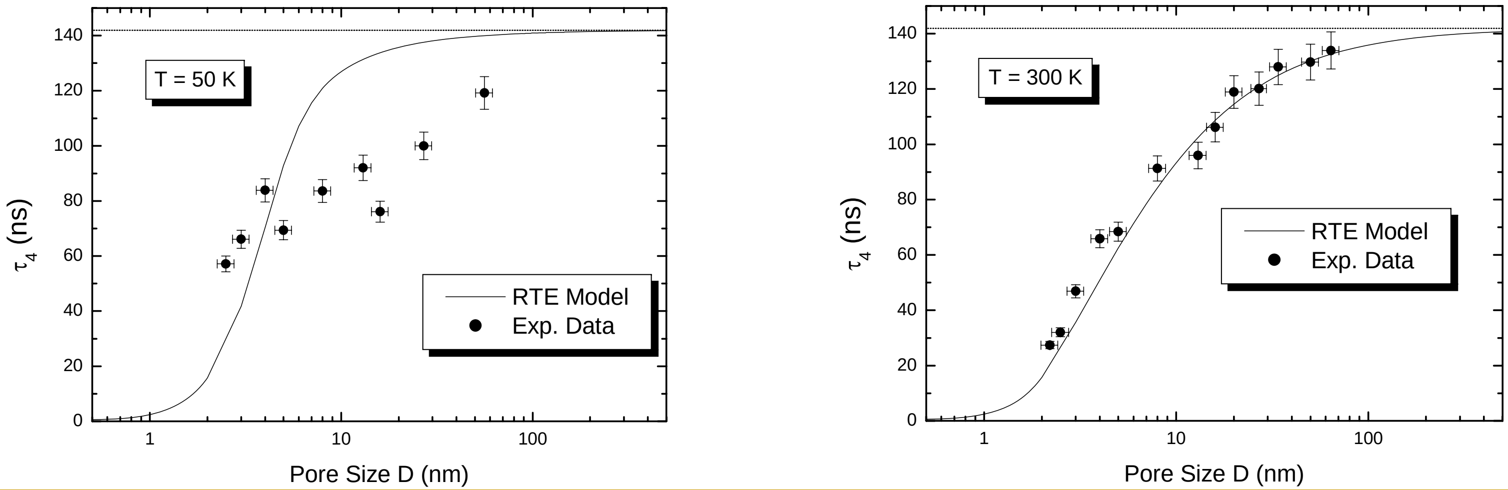
- **for D = 5 nm no temperature dependence to observe**
- **for D = 27 nm contrary behaviour to the RTE model**

Summary

- for T = 300 K general agreement to the RTE model -> at room temperature, PALS is a useful porosimetry tool!

- for T > 300 K still agreement to the RTE model.

- for low temperatures the measurements show disagreement to the RTE model



Summary – temperature dependence

- for small pores (D < 3 nm) the lifetime increases at low temperatures
- for D = 4 nm temperature dependence of lifetime fairly agrees to the RTE model
- for D = 5 nm no temperature dependence of the lifetime can be observed
- for large Pores (D > 8 nm) the lifetime decreases at low temperatures -> contrary behaviour to the model.

- Thanks for your patience!
- This talk as pdf?
- http://positron.physik.uni-halle.de