Enhanced decay and line broadening of 2P *ortho*-positronium inside silica pores

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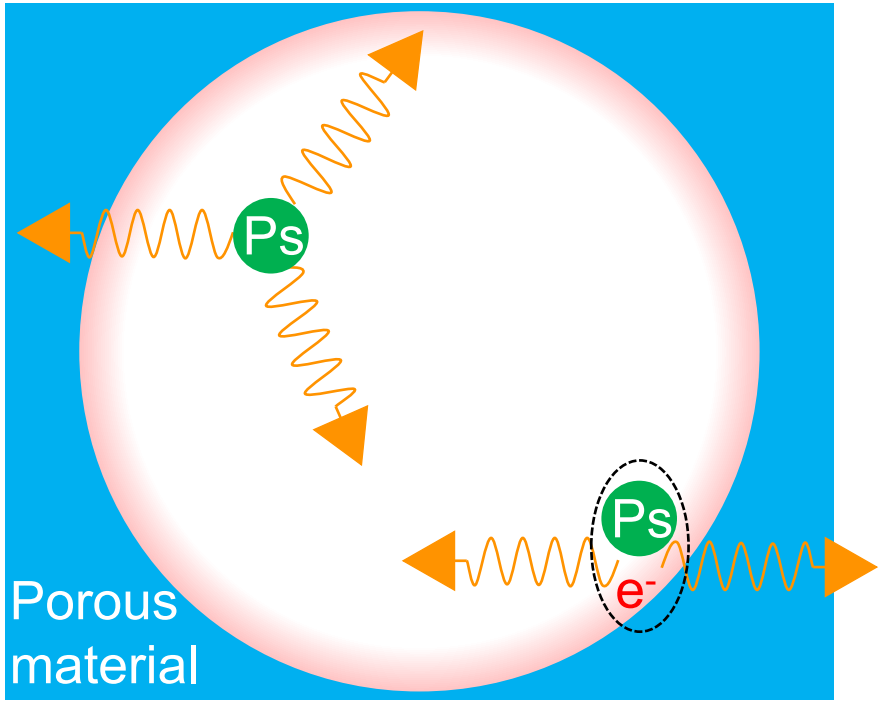
12.5th International Workshop on Positron and Positronium Chemistry
Online

Interaction of Ps with matters has make Ps useful probe

Pick-off decay

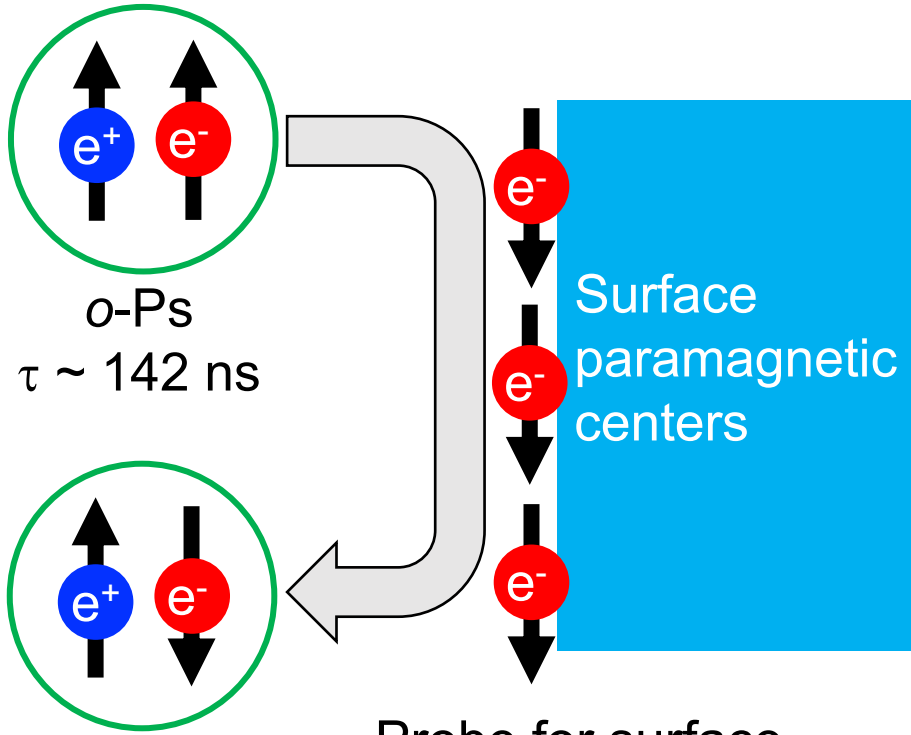
Decay of Ps with an electron on the material surface



Can inspect porous structure such as pore sizes and porosities

Spin-conversion

o-Ps's spin-flip to short-lived *p*-Ps by paramagnetic centers

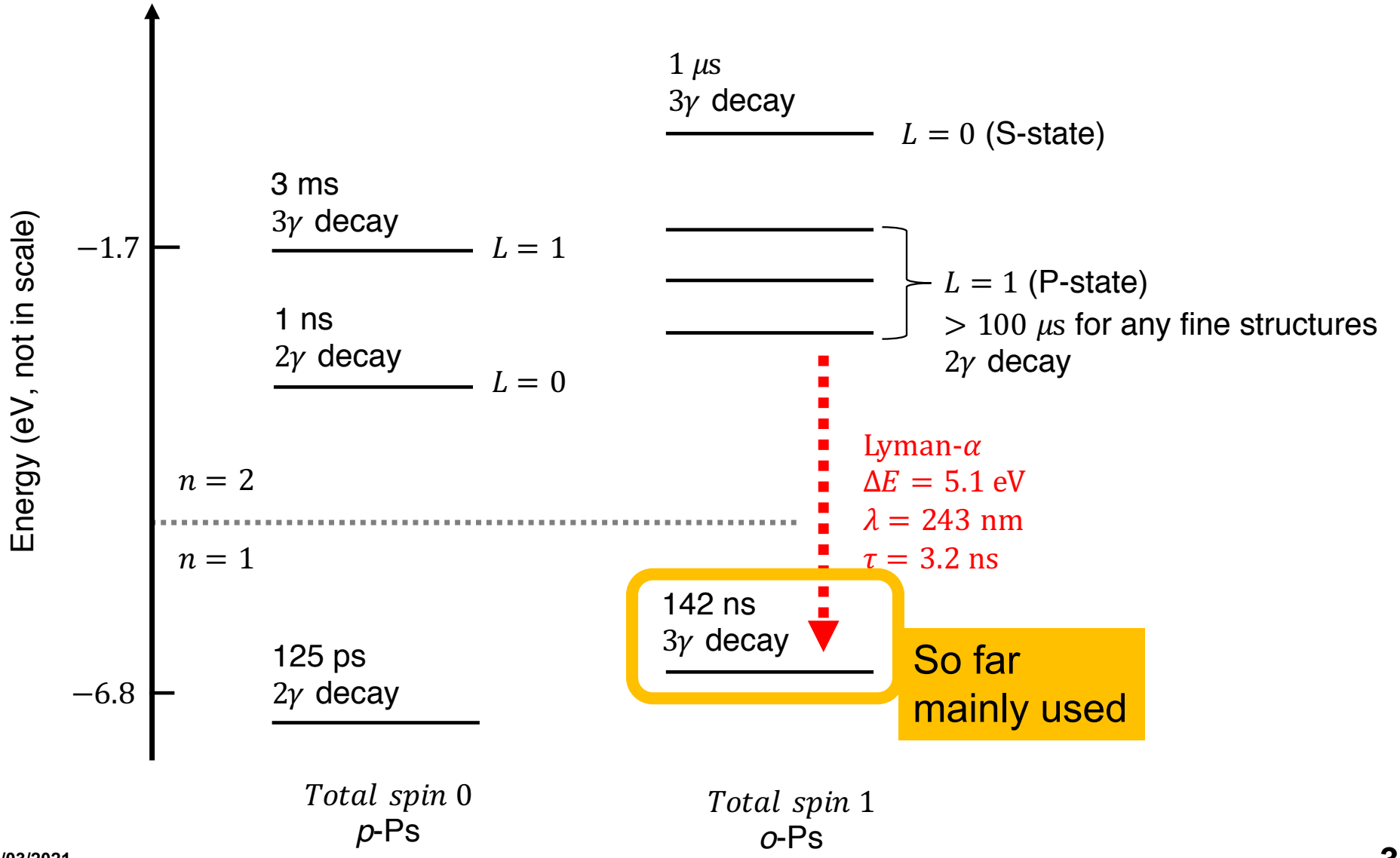


o-Ps
τ ~ 142 ns

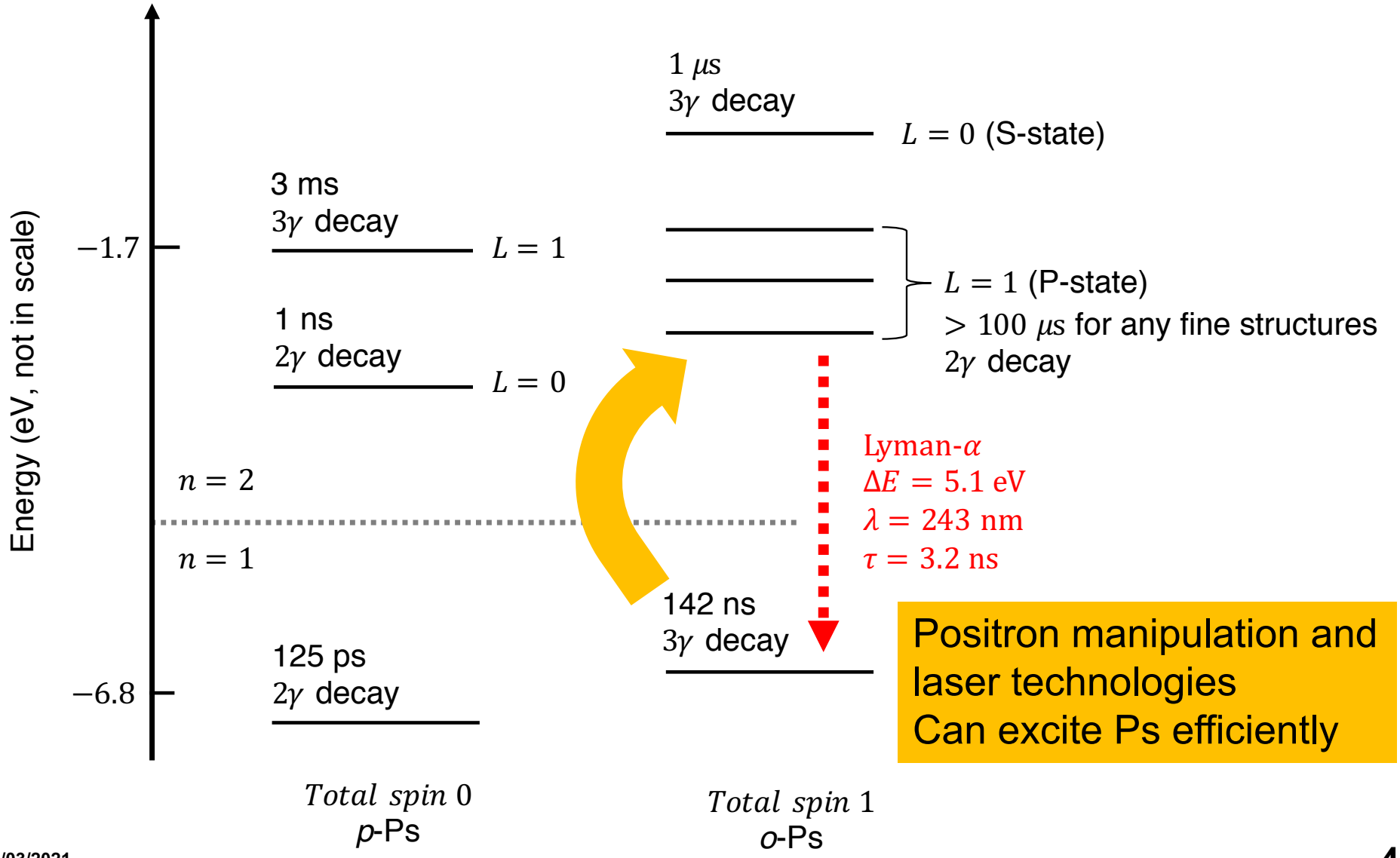
p-Ps
τ ~ 125 ps

Probe for surface unpaired spin

Ps in its excited state exhibits another face



Ps in its excited state exhibits another face

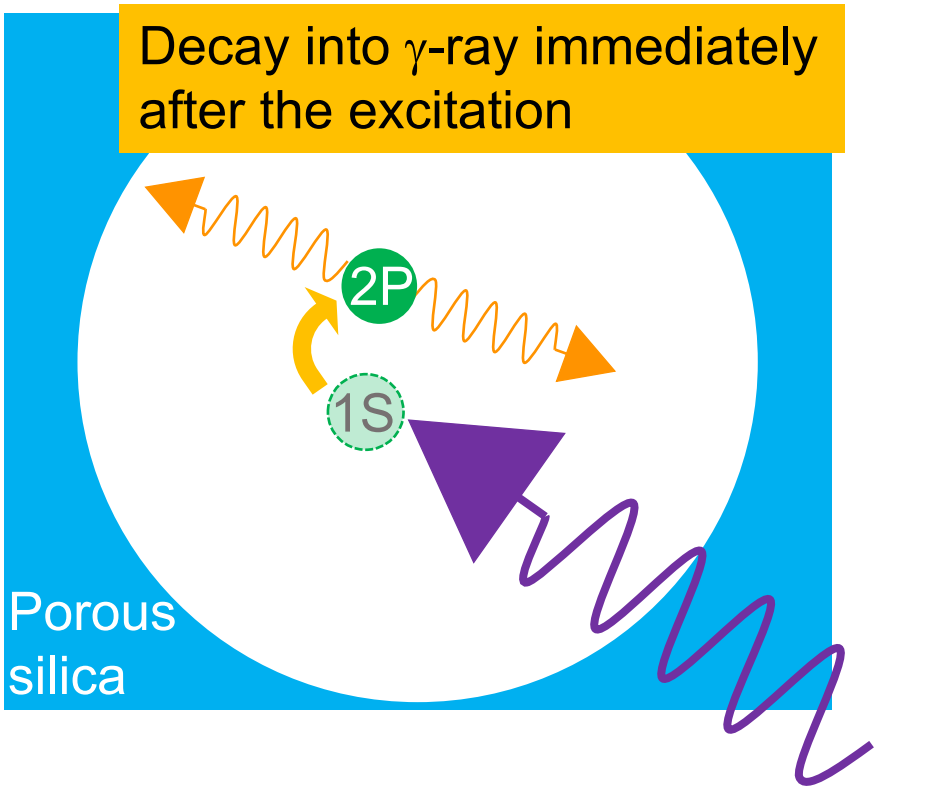


Positron manipulation and laser technologies
Can excite Ps efficiently

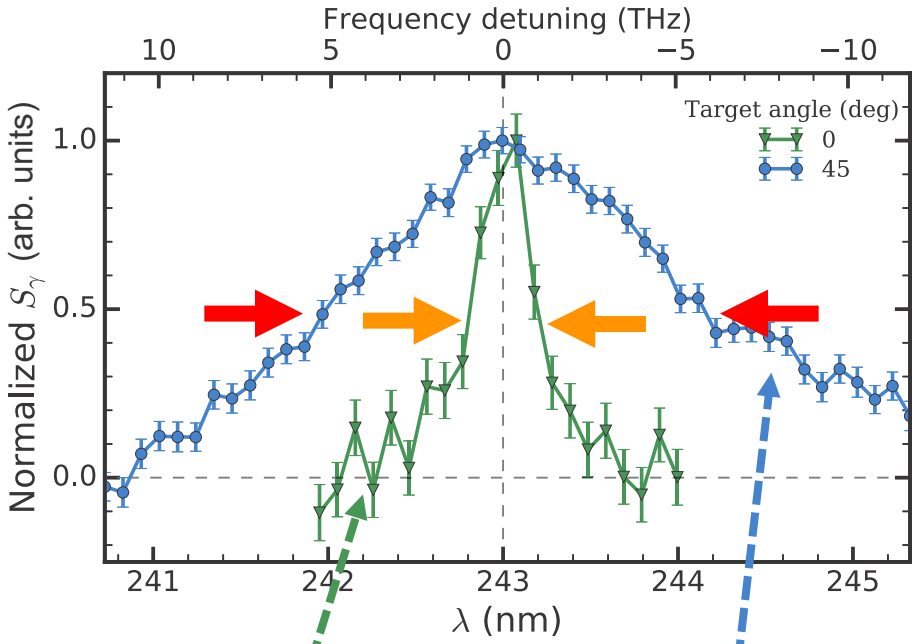
Unexplained rapid decay and line broadening have been discovered

Cooper *et al.*, Phys. Rev. B **97**, 205302 (2018) reported unexplained results in excitation into the 2P state

Rapid decay after excitation



Line broadening



Cooper *et al.*, Phys. Rev. B **97**, 205302 (2018)

Doppler width for Ps in vacuum

Much wider reason?

To be a new way as material probe and Ps cooling for fundamental study

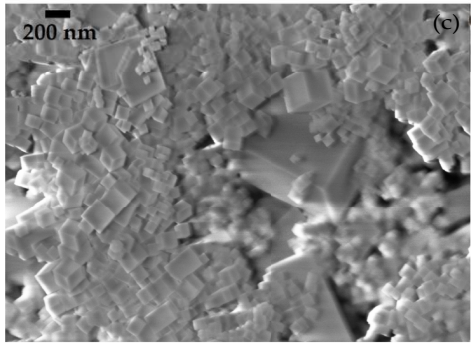
By understanding interactions between 2P-Ps and materials :

1. Use 2P-Ps as a new probe of material structure

In **some materials**, the unexplained result was **not** observed

Another porous silica

Cassidy et al.,
Phys. Rev. Lett. 106, 023401 (2011)



MgO smoke

Gurung *et al.*, Phys. Rev. A 101, 012701 (2020)

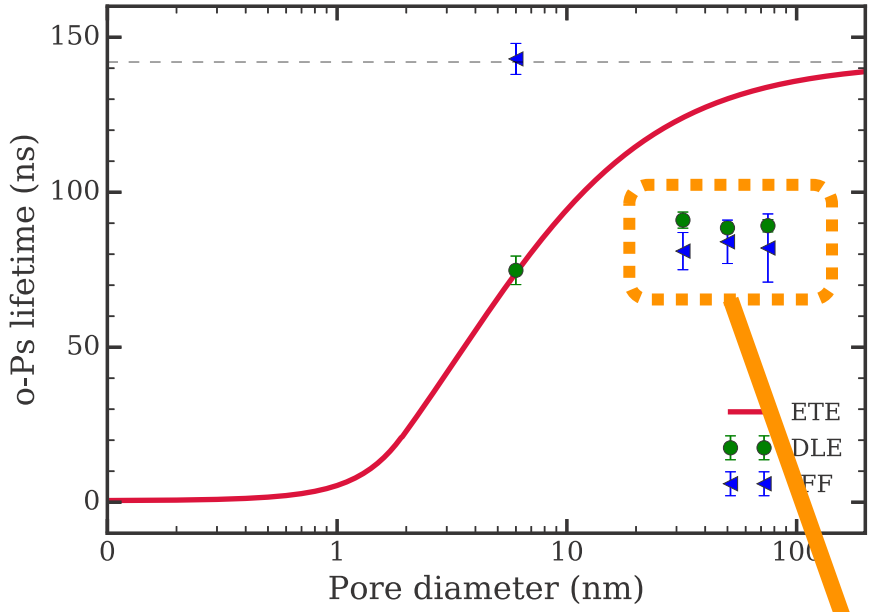
2P-Ps should respond some structures in material. **Probe it!**

2. Enable rapid Ps cooling combining with laser cooling

- Laser cooling on Ps in porous silica will be effective
  Shu *et al.*, J. Phys. B: At. Mol. Opt. Phys. **49**, 104001 (2016)
- Breakthrough to Ps-BEC, precise spectroscopy
- Please refer talks by A. Ishida, R. Uozumi, Y. Tajima on August 31
- Suppressing the unexplained results is necessary

Contaminants are trapping Ps?

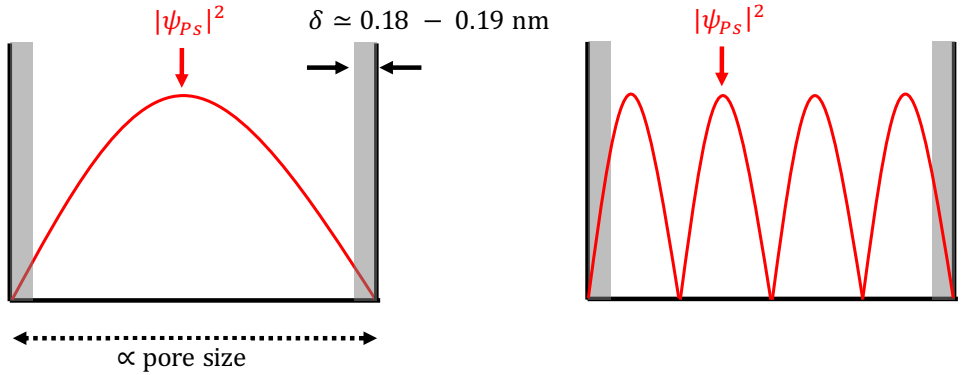
In Cooper *et al.*, Phys. Rev. B **97**, 205302 (2018), contaminants on pore surface were suspected



Cooper *et al.*, Phys. Rev. B **97**, 205302 (2018)

Not valid?

- In their nano pores, 1S o-Ps lifetimes were much shorter than predictions by RTE model



Predicts pick-off decay rate quantitatively

- Ps has the wavefunction in infinite squared well
- Ps has a large annihilation rate in the vicinity of the surface

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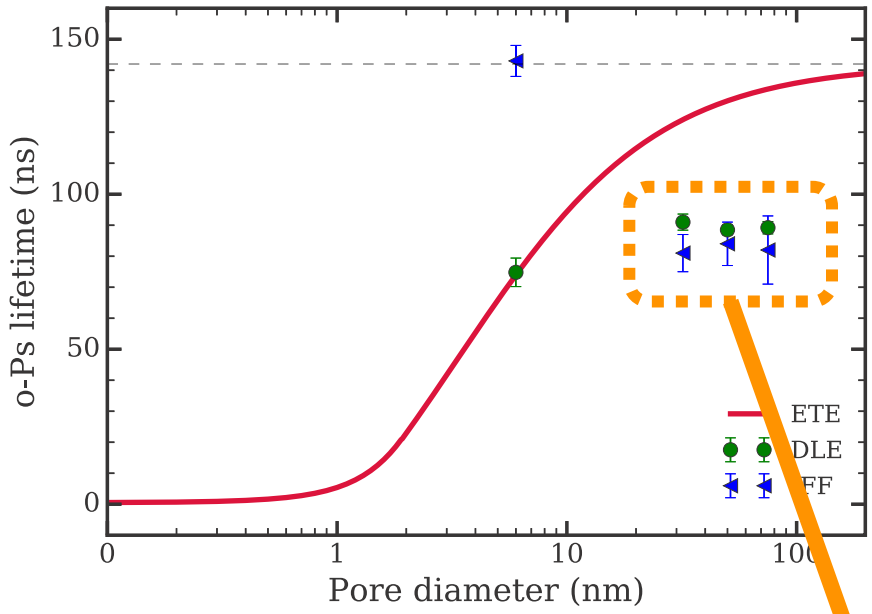

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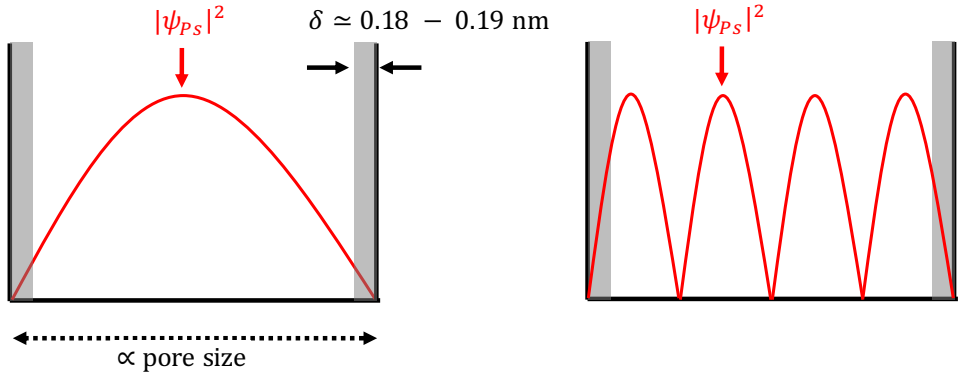


Not valid?

Predicts pick-off decay rate quantitatively

- Ps has the wavefunction in infinite

1. test this hypothesis
2. study basic properties of these phenomena

tion rate in the vicinity of the surface

Experiments at KEK-SPF

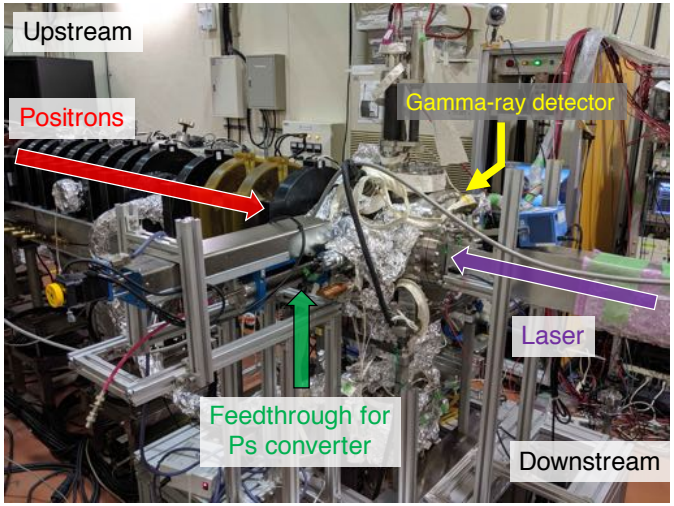


Figure 2.1: Picture of the experimental setup.

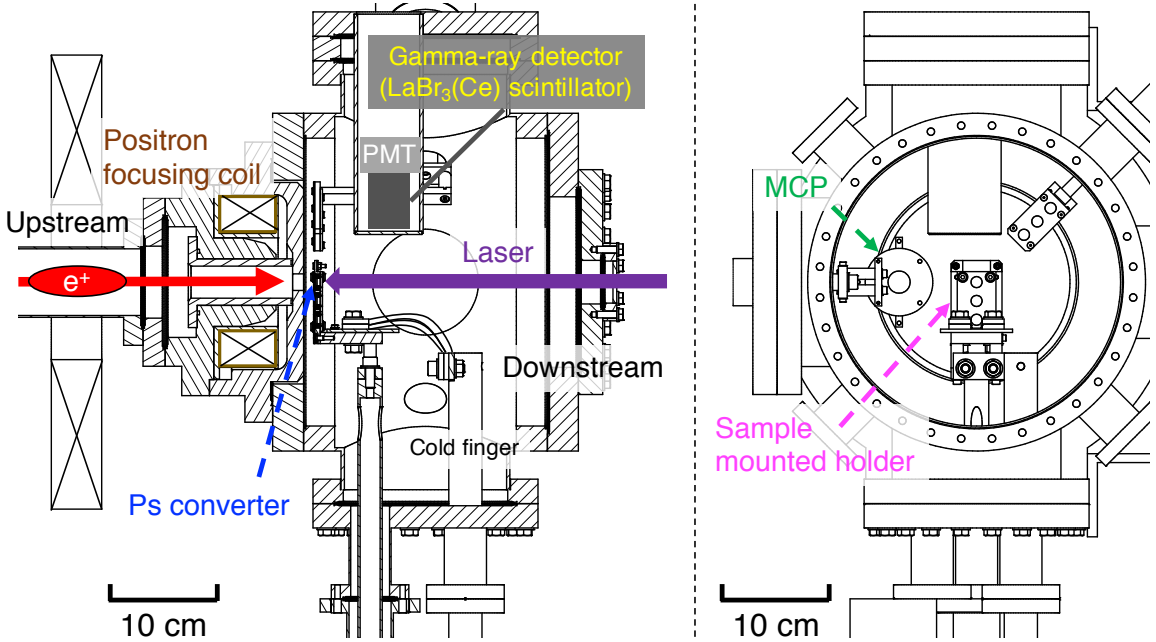
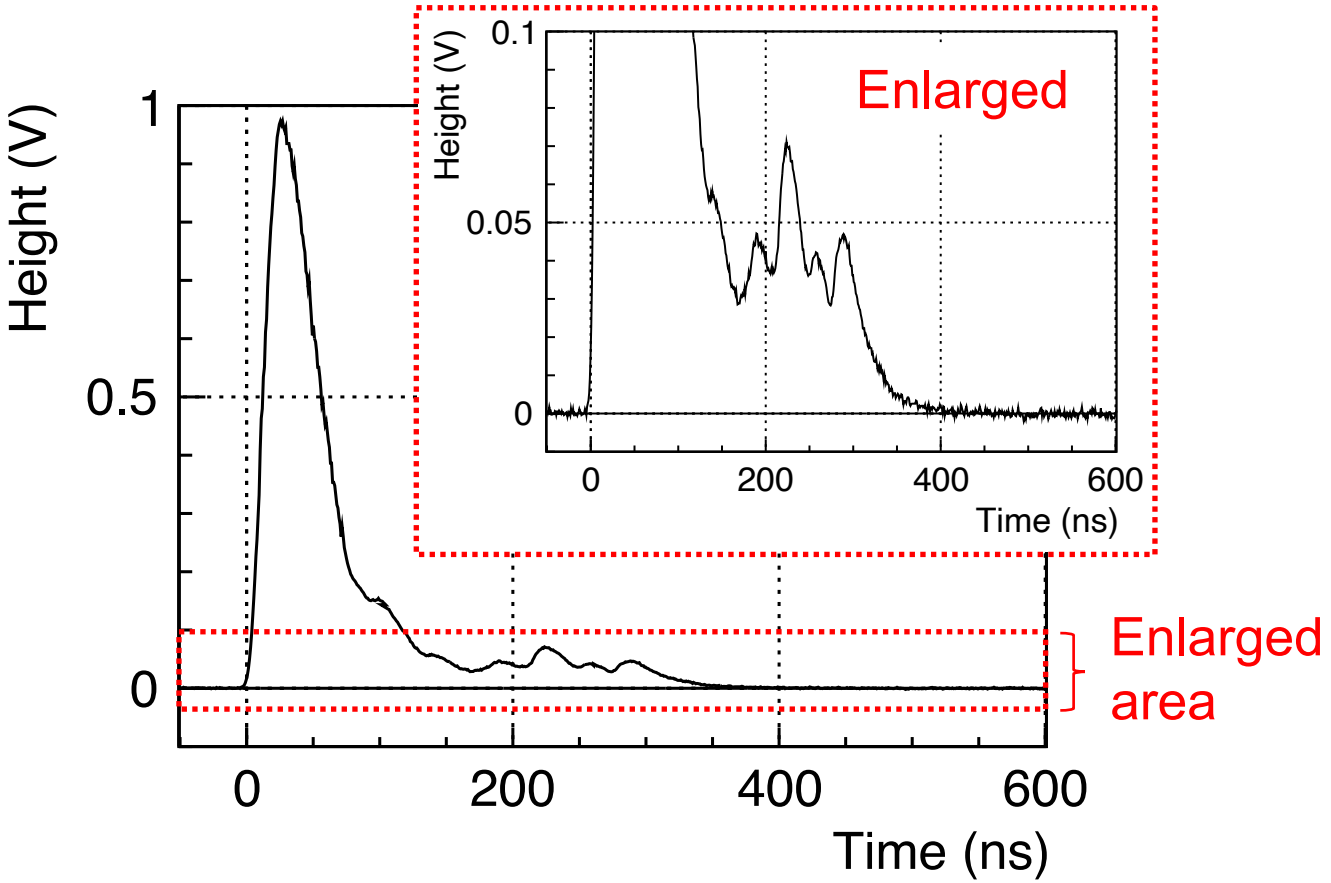


Figure 2.2: Drawings of the experimental chamber to excite Ps confined in the Ps converter.

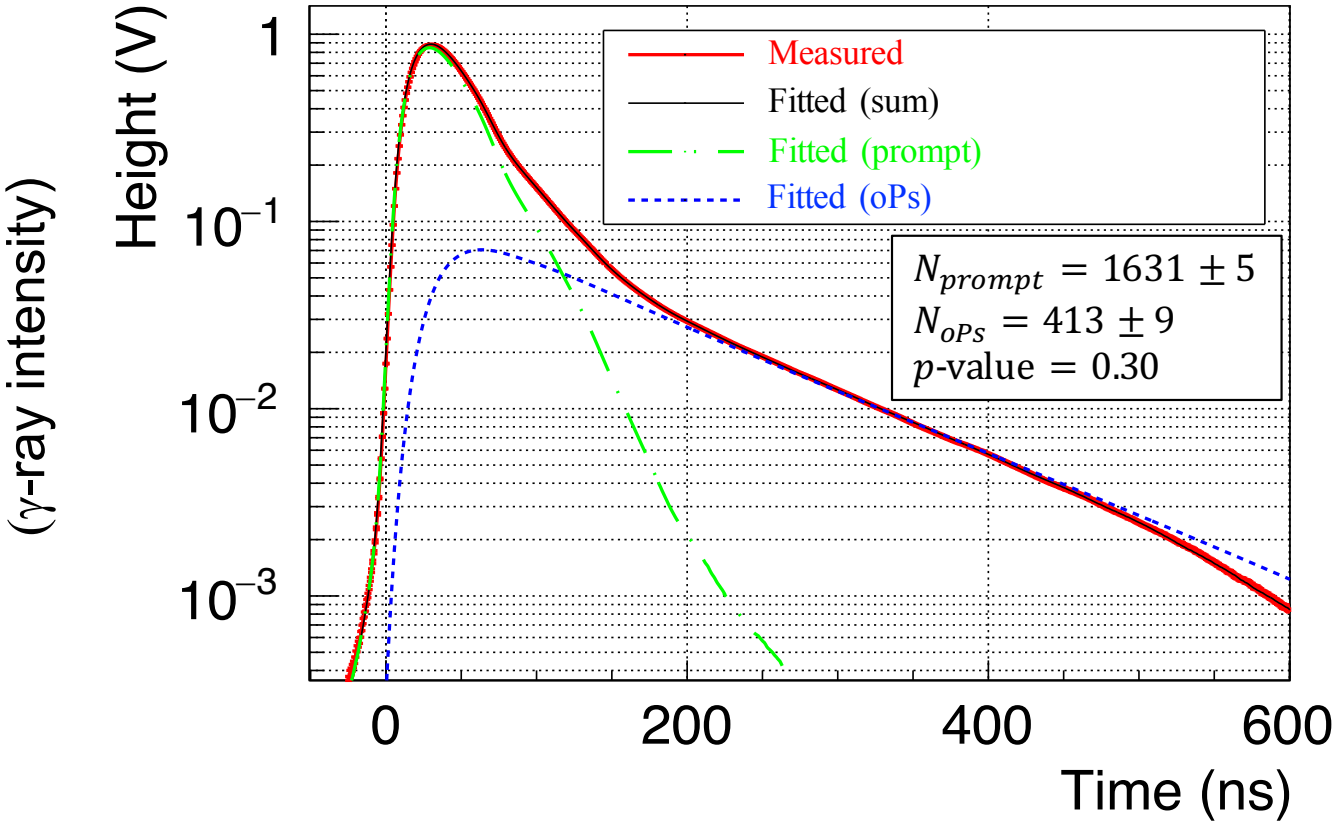
- KEK-SPF can provide bunched slow positron beam
- Silica aerogel was used as a Ps converter and trap
- UV nano-second pulsed laser was synchronized

Bunched positrons produce a lot of γ-rays detected with pileup



Typical PMT waveform which detected γ-rays

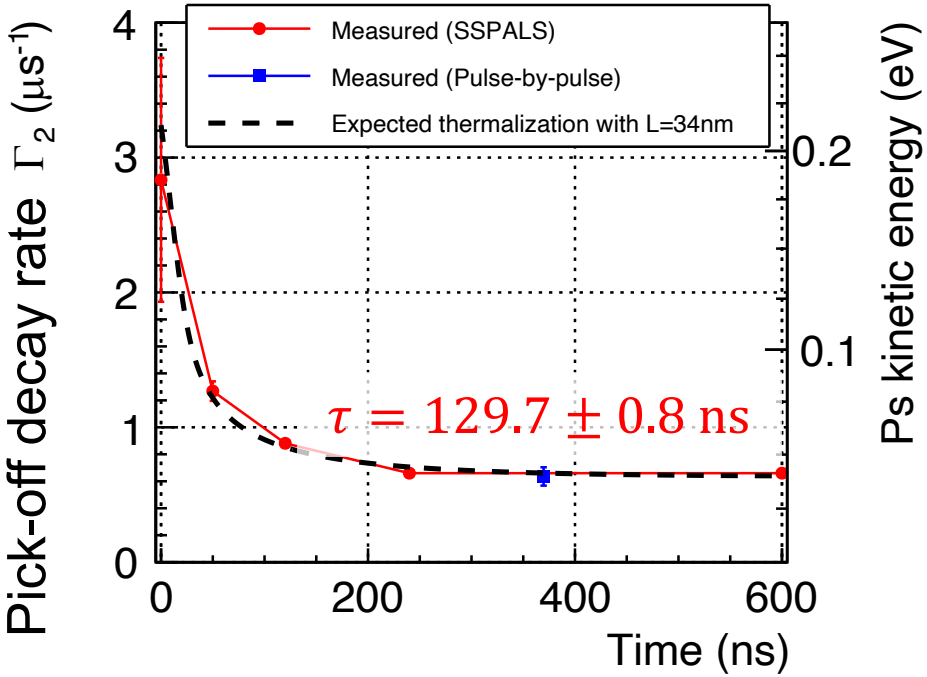
We adopt the SSPALS method



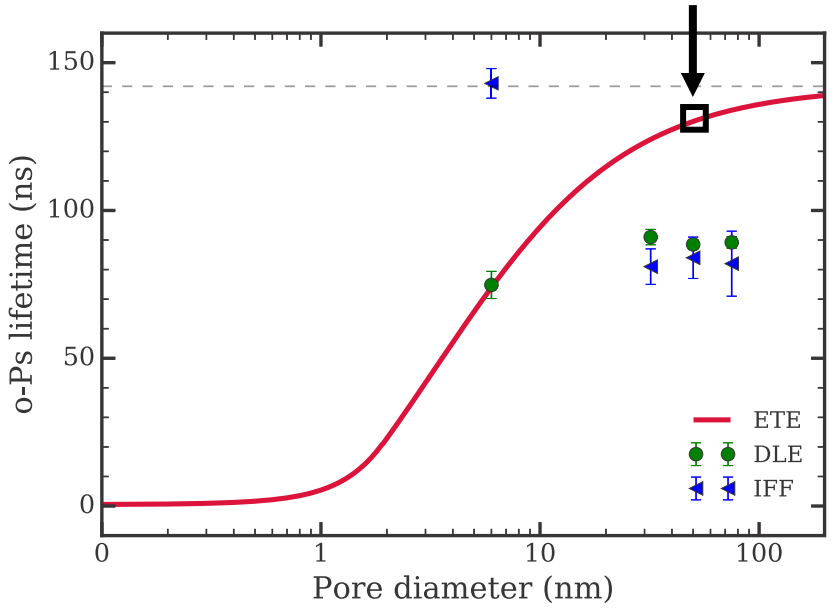
Measured average waveform and fitted function

Averaged waveforms over bunches were analyzed with a modeled function including Ps lifetime and intensity

Lifetime of 1S *o*-Ps agreed with the RTE model prediction



In our aerogel :
D = 50 nm, τ ~ 130 ns



Cooper *et al.*, Phys. Rev. B **97**, 205302 (2018)

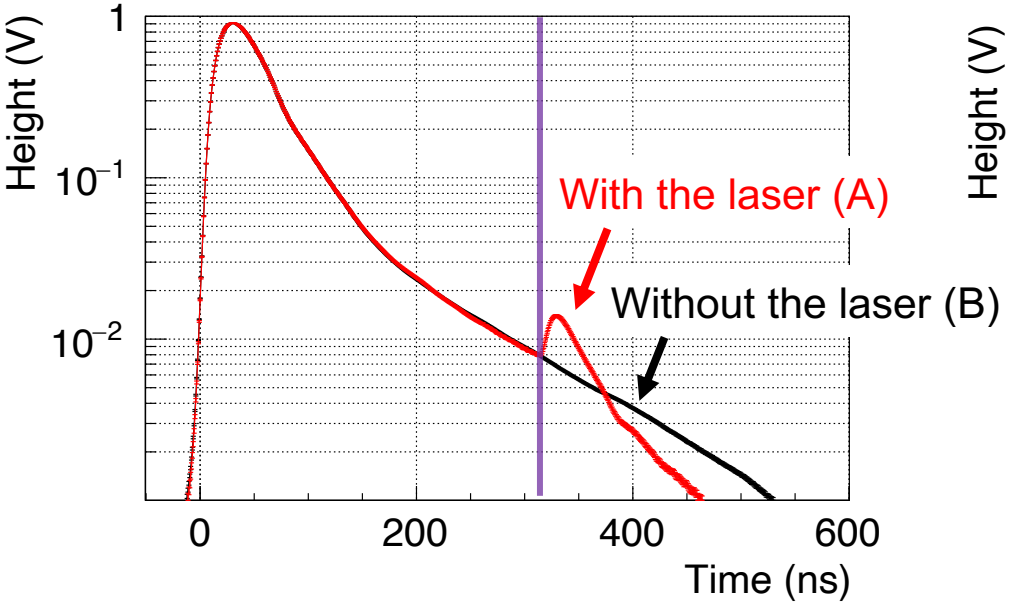
Thermalized lifetime was

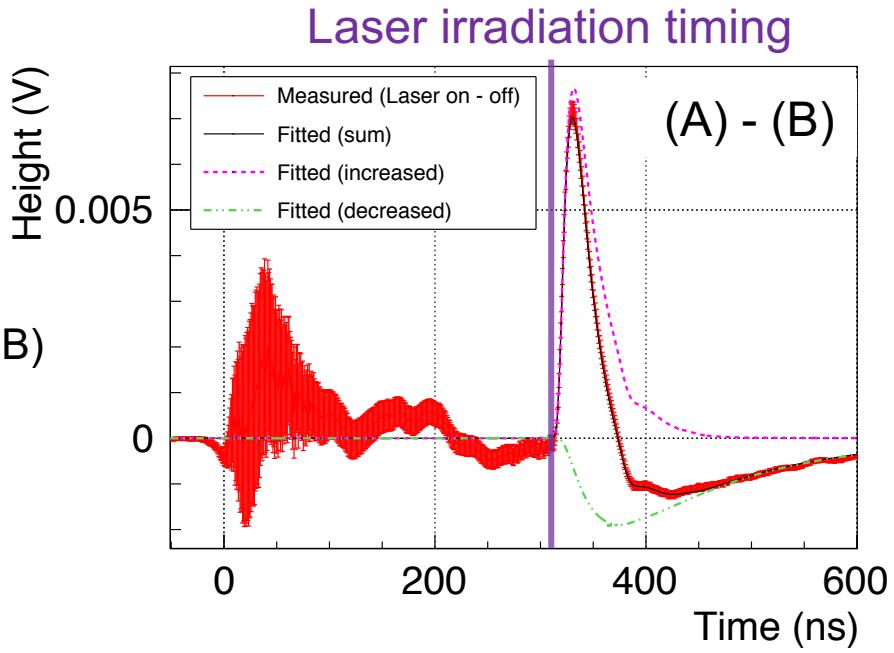
$$\tau = 129.7 \pm 0.8 \text{ ns}$$

Agreed with other methods including Bulk-PALS measurement

Contaminants are much less than the previous work

Even with less contaminants, excitation induced the enhance decay





Waveform averages with / without laser

Difference by laser irradiation fitted by the following model

Laser excitation triggered enhanced decay into γ-rays

Quantitatively model the enhanced decay

To quantitatively study the unexplained result, we modeled the enhanced decay by the optical Bloch equation

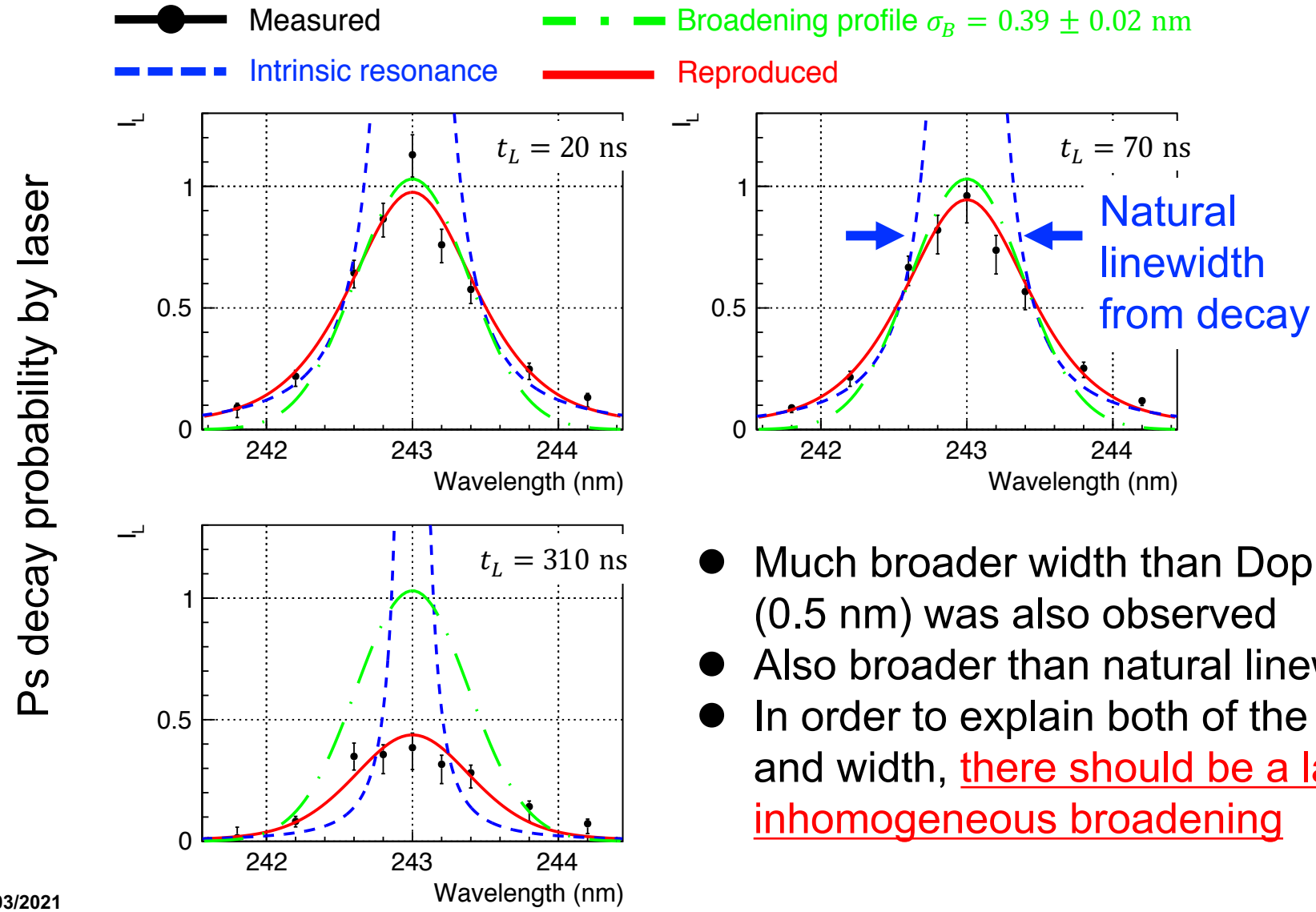
Ps wavefunction : $|\Psi(t)\rangle = c_1(t)|1S\rangle + c_2(t)|2P\rangle, \qquad \rho_{ij} = c_i^* c_j$

$$\begin{cases} \dfrac{d}{dt}\rho_{11} = \dfrac{i}{2}\Omega(\rho_{12} - \rho_{21}) - \Gamma_1\rho_{11} + (\Gamma_{sp} + P_1\Gamma_2)\rho_{22}, \\ \dfrac{d}{dt}\rho_{12} = \dfrac{d}{dt}\rho_{21}^* = \dfrac{i}{2}\Omega(\rho_{11} - \rho_{22}) + \left(i(\omega_{21} - \omega_L) - \dfrac{1}{2}(\Gamma_1 + \Gamma_{sp} + \Gamma_2)\right)\rho_{12}, \\ \dfrac{d}{dt}\rho_{22} = -\dfrac{i}{2}\Omega(\rho_{12} - \rho_{21}) - (\Gamma_{sp} + \Gamma_2)\rho_{22}. \end{cases}$$

Γ_2 is the enhanced decay rate of 2P Ps
By inputting laser parameters (pulse energy, wavelength),
we can calculate **Ps decay probability**

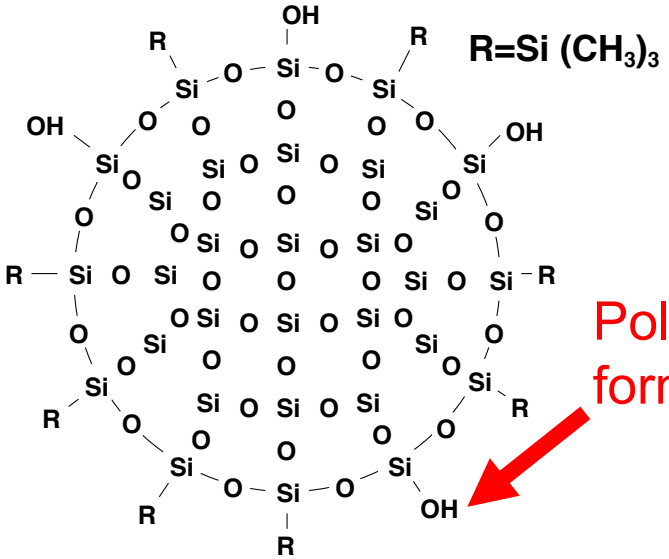
Estimated by fitting measured waveform

There should be inhomogeneous broadening



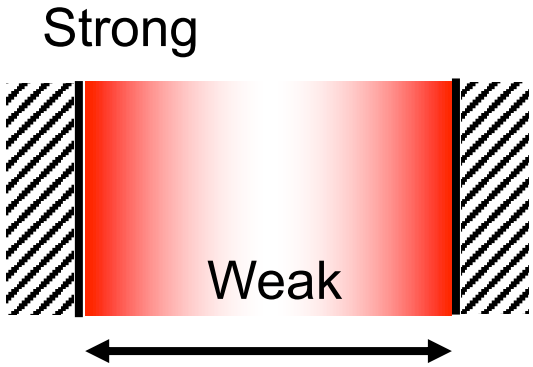
- Much broader width than Doppler (0.5 nm) was also observed
- Also broader than natural linewidth
- In order to explain both of the prob. and width, there should be a large inhomogeneous broadening

Distribution of Stark shift can cause inhomogeneous broadening



Structural diagram of silica grain

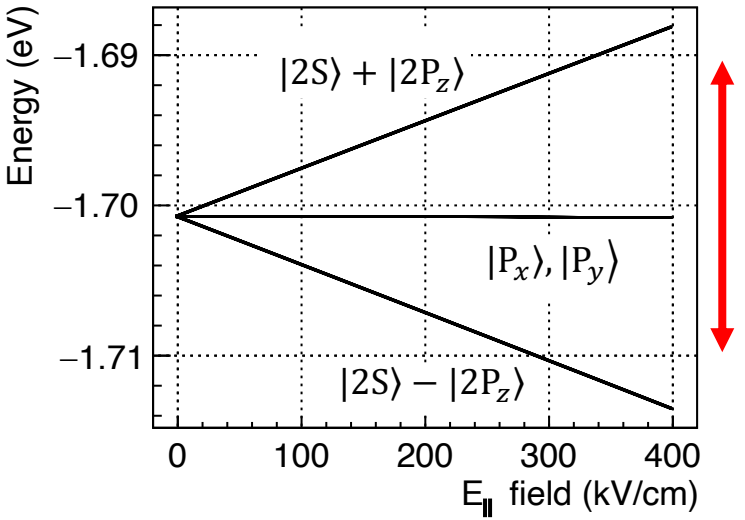
Y. Kataoka, PhD thesis (2007)



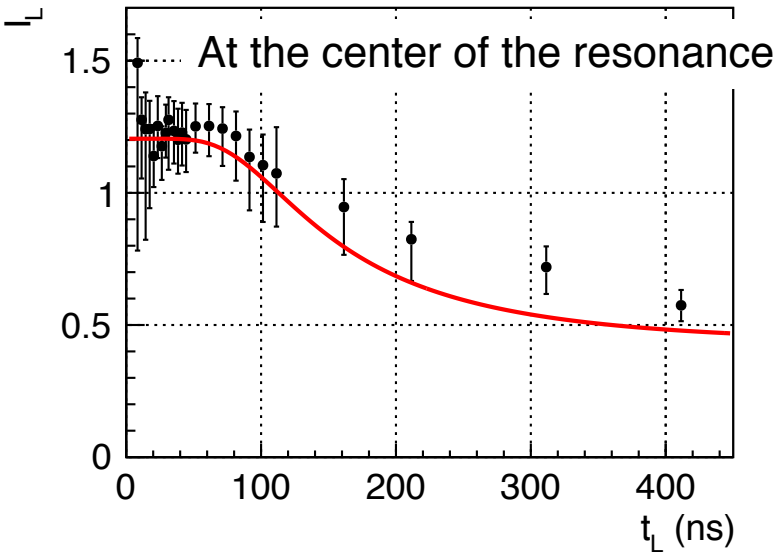
Field strength distribute against distance from surface

- Electric field shifts Ps energy, then resonant wavelength
- Field distribution cause broadening
- Field inside aerogel pore can be strong enough to cause the observed broadening

0.02 eV = 1 nm of 243 nm

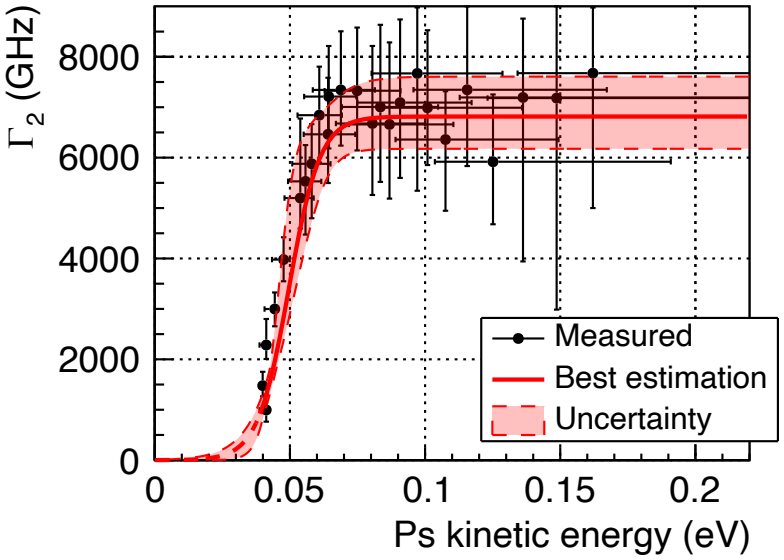


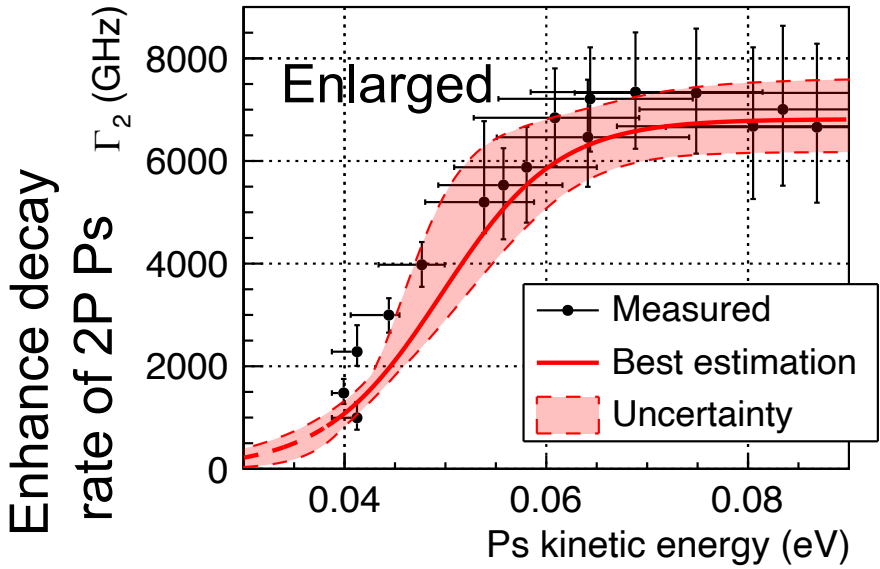
Enhanced decay rate had positive correlation with kinetic energy



Laser timing

By optical Bloch eq.

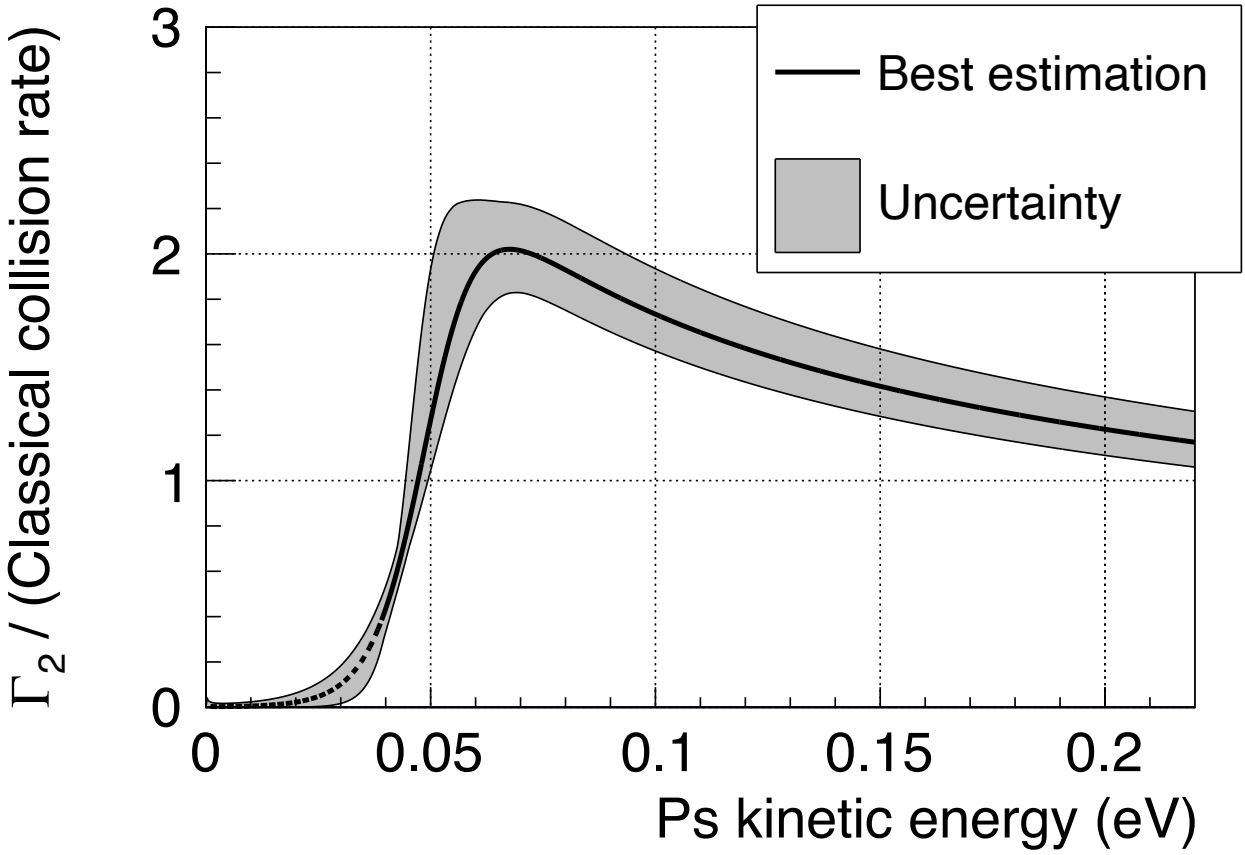




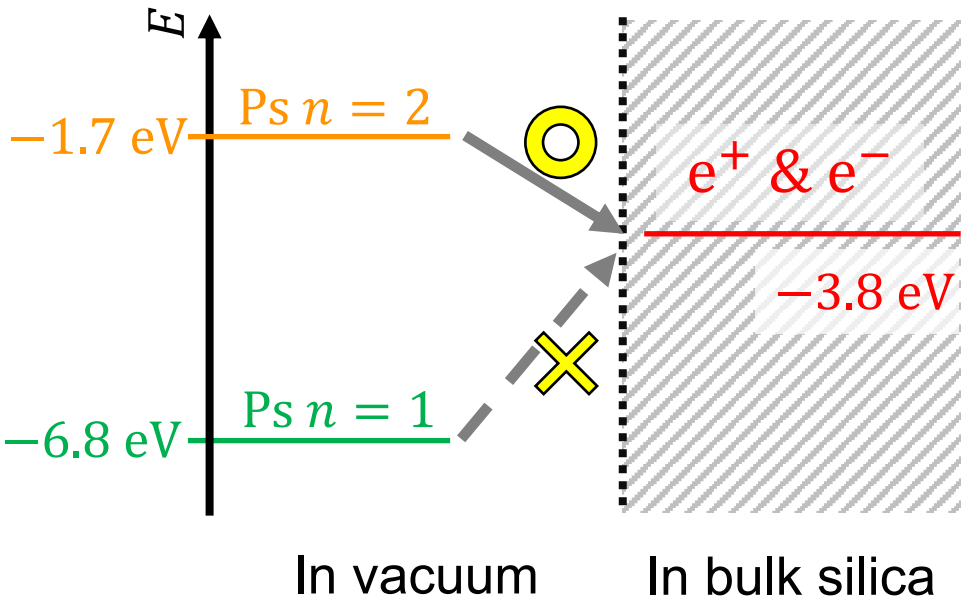
- Because Ps gradually slow down to aerogel temperature, energy dependence can be measured
- Enhanced decay rate in several THz varied with Ps kinetic energy

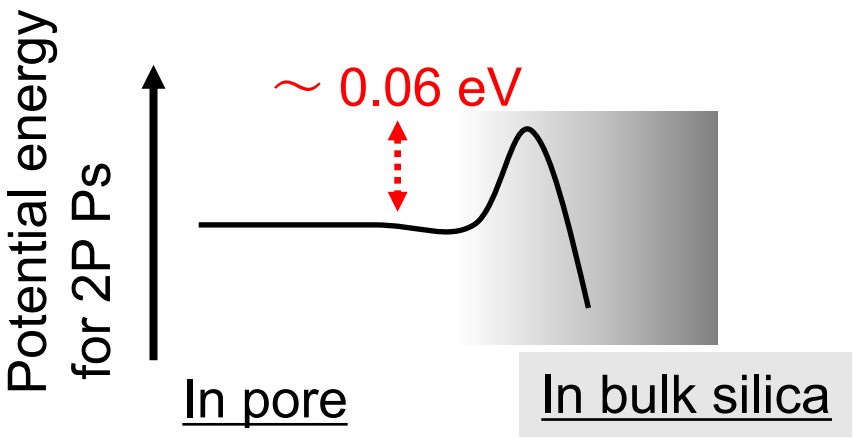
Decay rate was in the order of the classical collision rate

- Collision rate was estimated by kinetic energy and pore size
- Enhancement at 0.06 - 0.07 eV



Small binding energy for n=2 can lead large decay probability





For silica, Ps in n=2 would not be more stable than dissociated state in the bulk

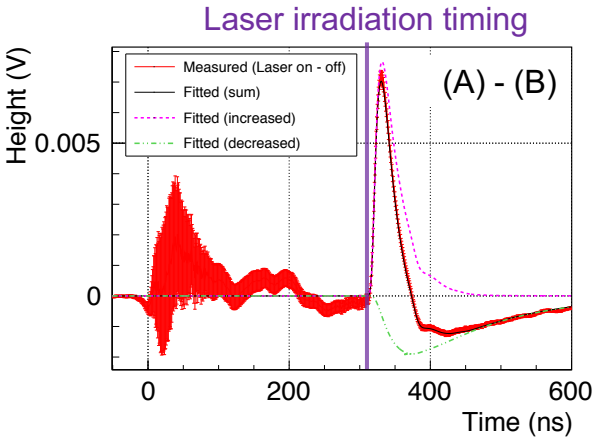
This would explain almost unity decay probability of 2P Ps

Energy dependence would have information on surface potential

More theoretical / experimental studies to test these models are necessary

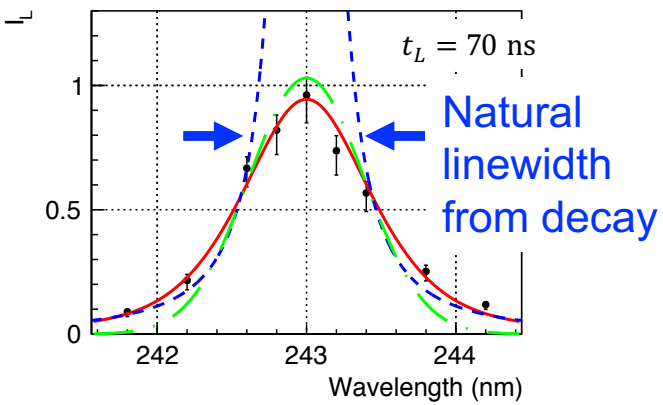
Summary

- Unexplained results for interaction between 2P Ps and materials were found. We conducted basic studies.
- Understanding them will lead to new way to use Ps as a material probe, and breakthrough on Ps cooling for fundamental physics study

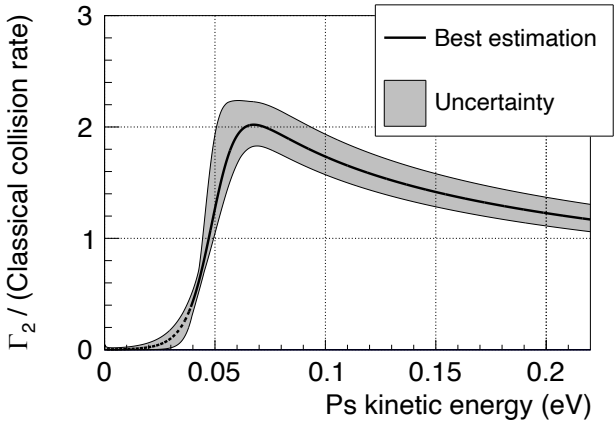


Difference by laser irradiation fitted by the following model

Enhanced decay was observed even in aerogel, which had less contaminants



Inhomogeneous broadening



Very efficient dissociation