Detection of light emission produced in the process of positronium formation

M. Pietrow [1], R. Zaleski [2], A. Wagner [2], P. Słomski [2], E. Hirschmann [2], R. Krause-Rehberg [2], M.O. Liedke [2], M. Butterling [2], D. Weinberger [2]

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September 23, 2019

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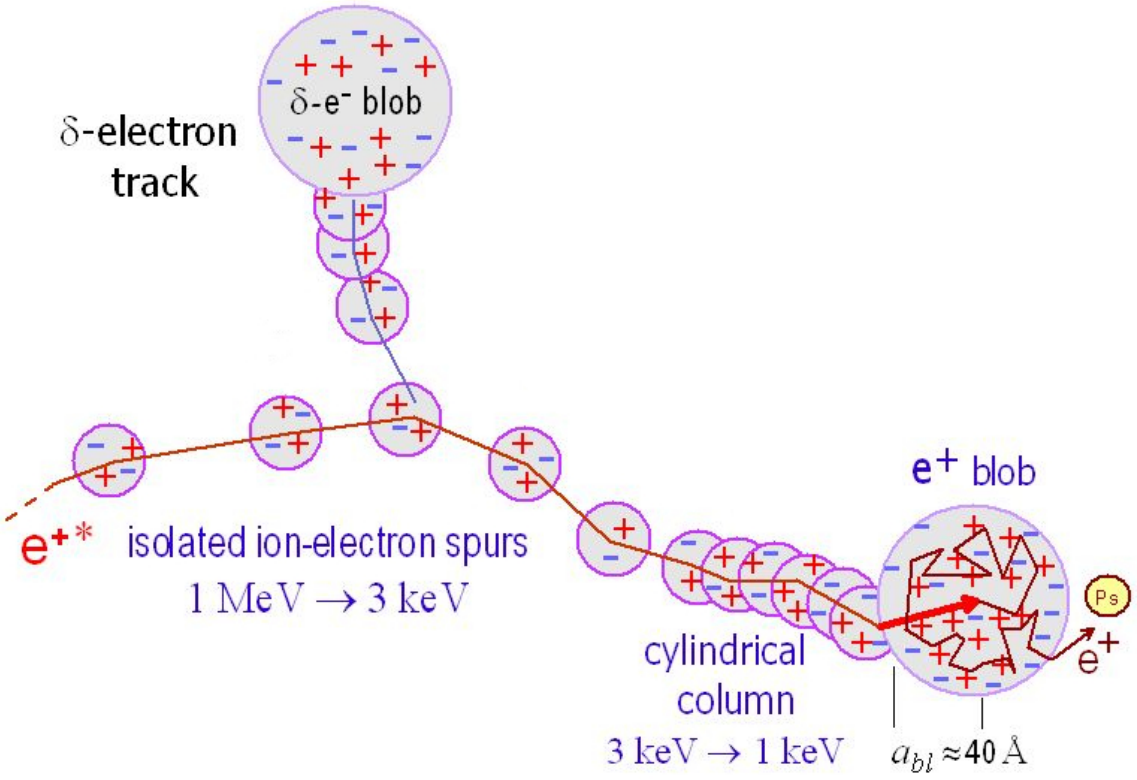
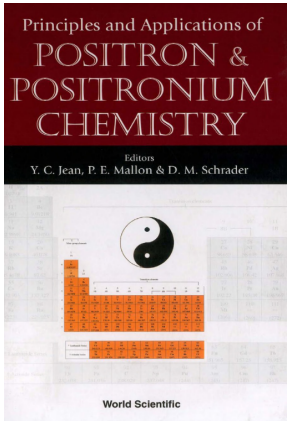
Ps formation model – the *blob* model



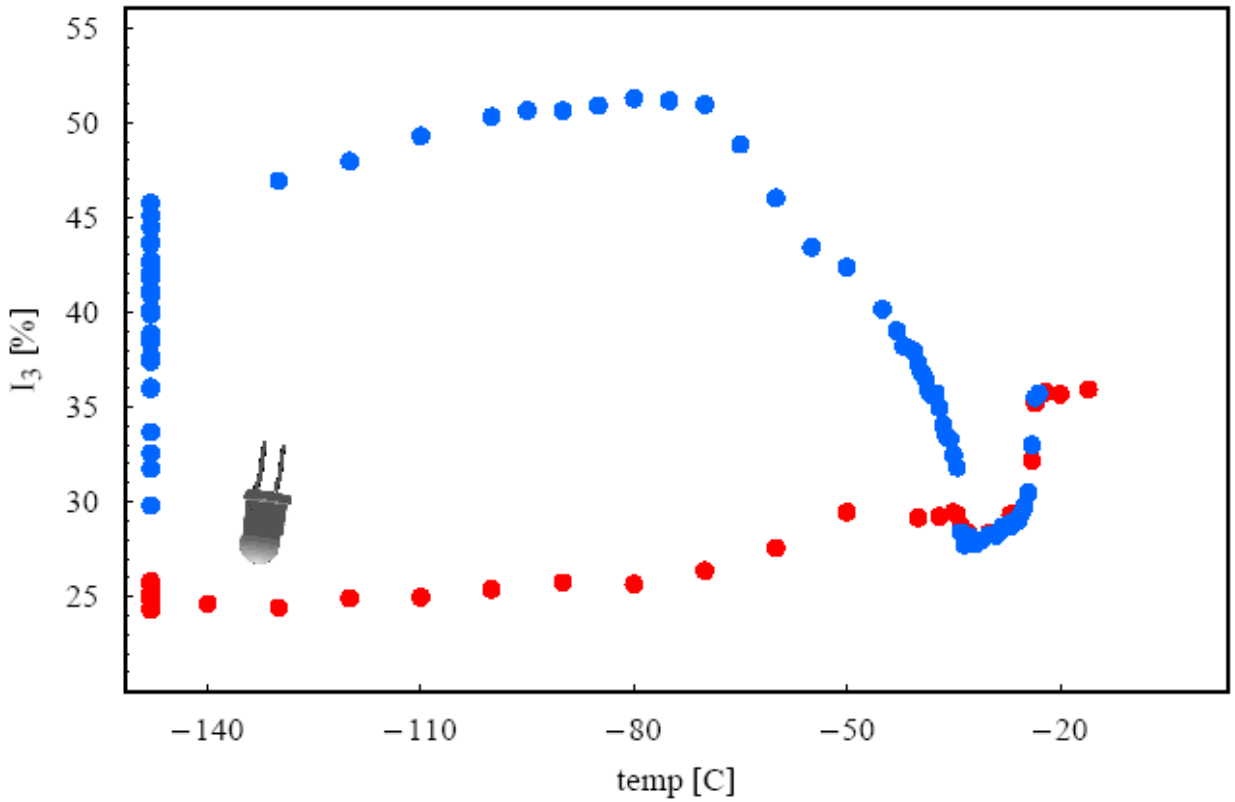


First ionization potential for alkanes is ∼10eV.

Ch. Cao, H. Yuan: J. Chem. Inf. Comput. Sci. 2002, 42, 667-672

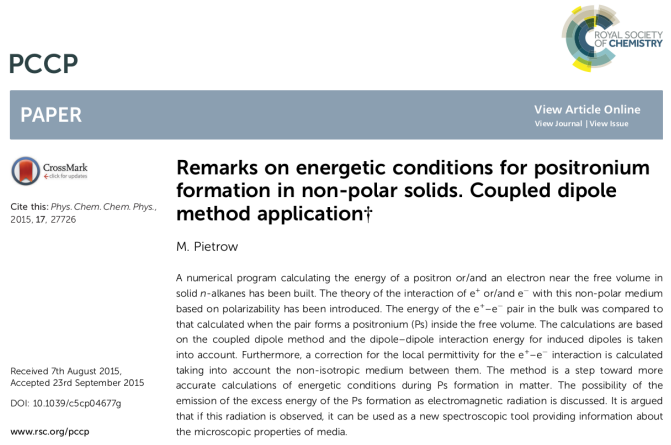
Ps formation model – trapped electrons

PALS experiment with alkane



energetic conditions for Ps formation

What happens in a bulk?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Cite this: *Phys. Chem. Chem. Phys.*, 2015, **17**, 27726

Remarks on energetic conditions for positronium formation in non-polar solids. Coupled dipole method application†

M. Pietrow

A numerical program calculating the energy of a positron or/and an electron near the free volume in solid *n*-alkanes has been built. The theory of the interaction of e^+ or/and e^- with this non-polar medium based on polarizability has been introduced. The energy of the e^+–e^- pair in the bulk was compared to that calculated when the pair forms a positronium (Ps) inside the free volume. The calculations are based on the coupled dipole method and the dipole–dipole interaction energy for induced dipoles is taken into account. Furthermore, a correction for the local permittivity for the e^+–e^- interaction is calculated taking into account the non-isotropic medium between them. The method is a step toward more accurate calculations of energetic conditions during Ps formation in matter. The possibility of the emission of the excess energy of the Ps formation as electromagnetic radiation is discussed. It is argued that if this radiation is observed, it can be used as a new spectroscopic tool providing information about the microscopic properties of media.

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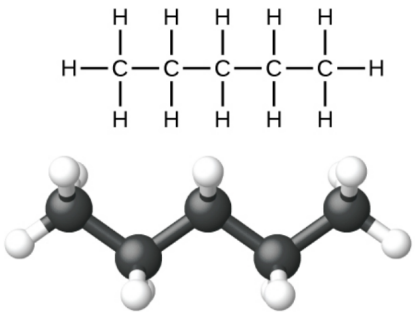
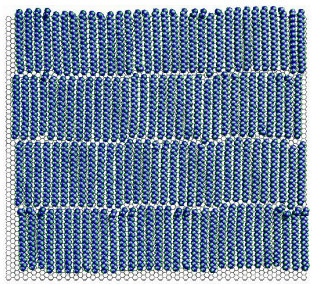
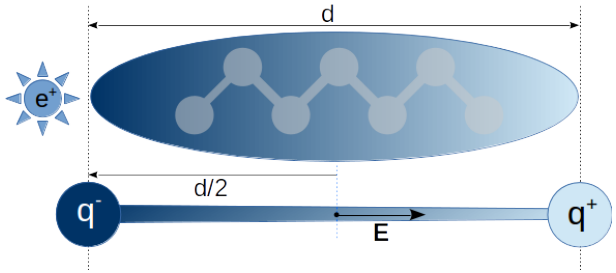
www.rsc.org/pccp

Let's take an n-alkane

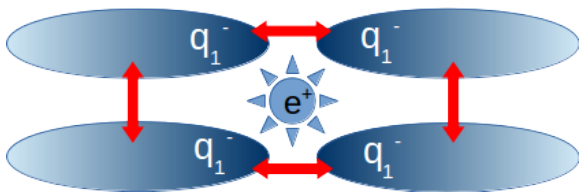




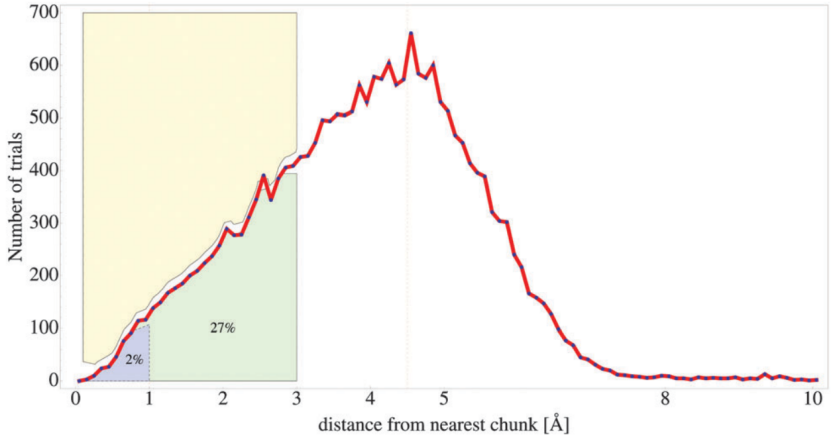


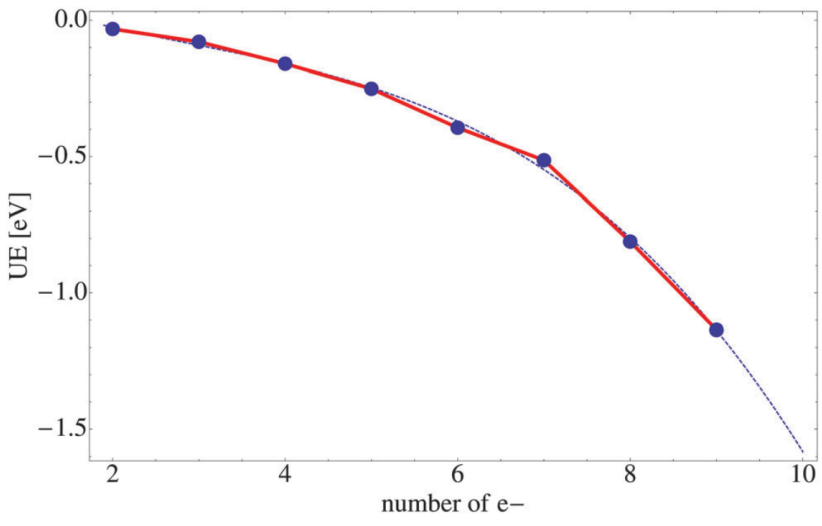
polarisability

$\alpha = 4.45038 \times 10^{-39} F \cdot m^2$

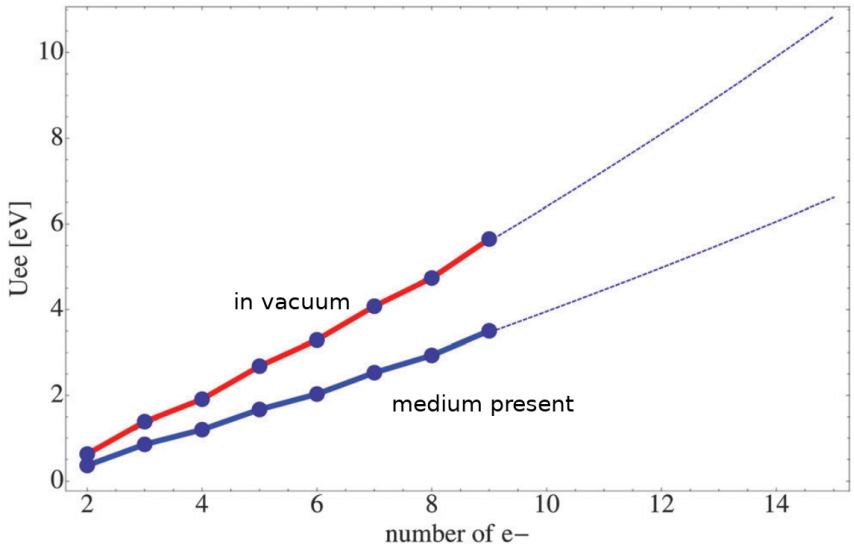


energetic conditions for Ps formation





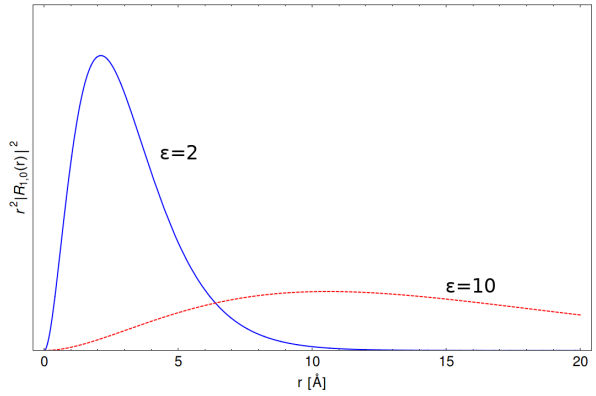
The energy of the interaction of quasi-free electrons with the environment.



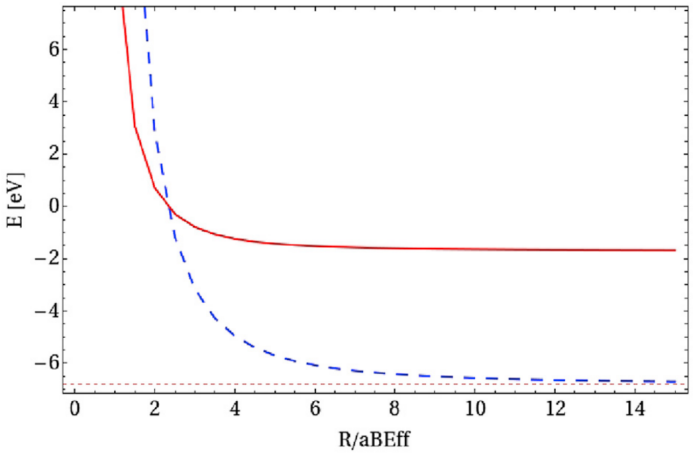
Interaction energy of an electron with other electrons as a function of the electron number

energetic conditions for Ps formation

What happens in a free volume?



radius of the exiton-like Ps in a bulk



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Application of the theory of excitons to study the formation of positronium and optical transition in matter

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Keywords:
Positronium formation
Exciton in quantum dot
Positron spectroscopy

ABSTRACT

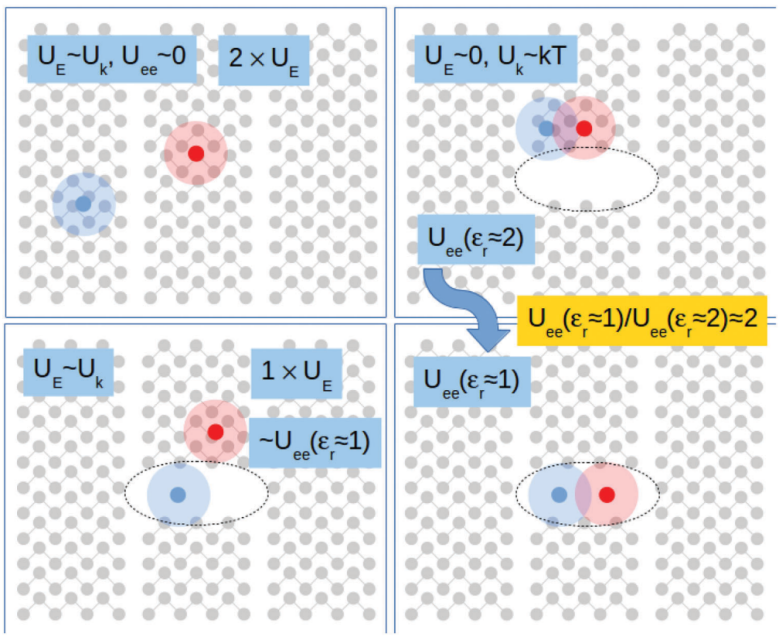
Considerable similarity between a positronium atom and an exciton in a quantum dot is indicated. Following this, we apply the calculation regime from the theory of excitons to describe an electron – positron pair near a free volume in matter where the positronium is created. It is shown that, in general, this actual confined state cannot be equated to a classical atom in vacuum.

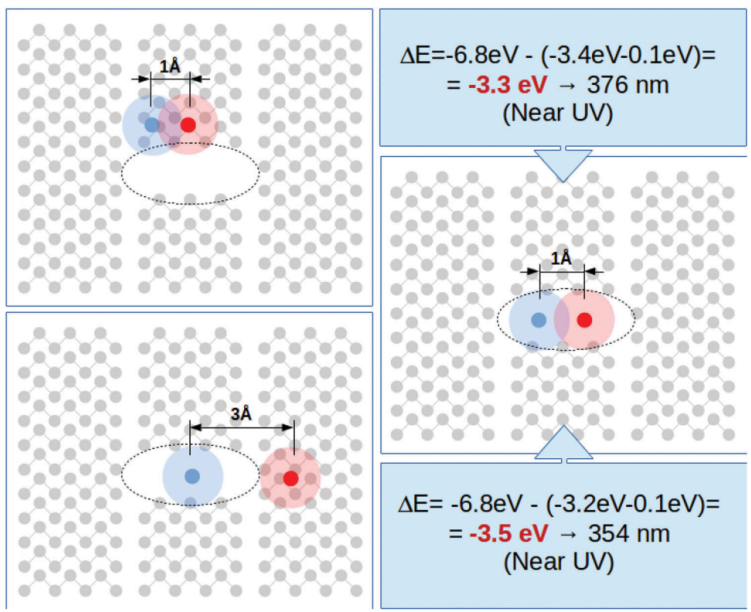
Besides the release of the energy of the pair in the phononic way during the Ps formation, we analyse the possibility of photonic deexcitation and show the way of calculation of its probability. The optical transitions speculated here are dependent on the electronic properties of the matter and, if detected, could allow improving experimental studies of solid matter properties with positron techniques.

energetic conditions for Ps formation

- Even for large radii of a free volume $e^+ - e^-$ polarizes molecules that affect its energy,
- The 'atom' is 'free' for $R/a_B^{\mathrm{eff}} \gtrsim 4$.

So, what is an energetic excess?





Where the energy excess is deposited? Phonons

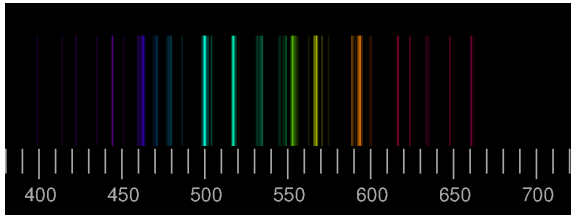
- The energy is too small to kick out the valece e^-
- q-neutral q-Ps loosely interacts with other q-free e^-

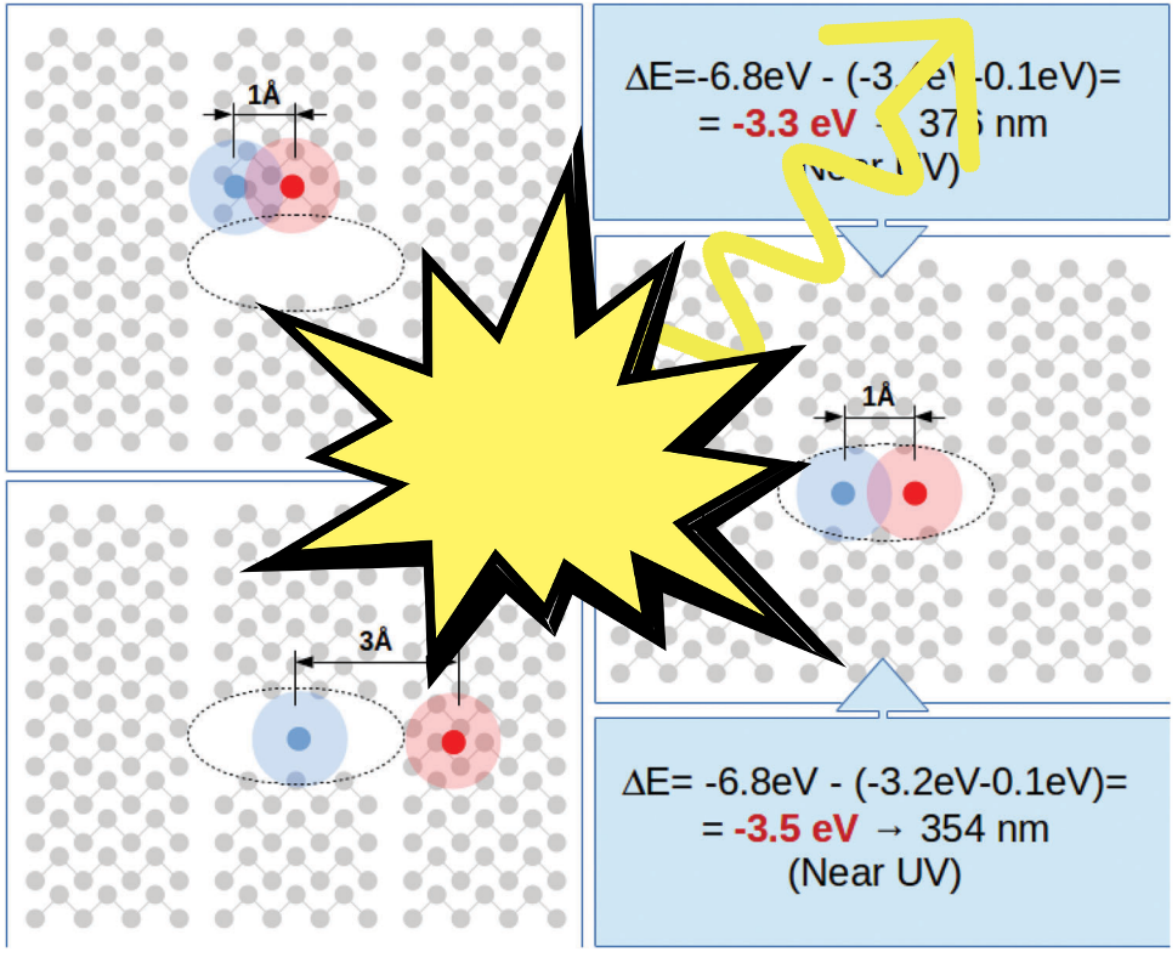
Phonons are not obvious in this case

- For phonons in crystals, the typical time scale is 10^{-12}s and energy scale is meV.
- The typical melting transition enthalpy is 40kJ/mol for an alkane. This gives 0.4eV energy per one molecule. Here, the energy of some eV is injected into the surrounding of the free volume (a few molecules).
- A release of some electron volts energy via mechanical oscillations in a few nanoseconds should produce a local melting (a collapse a free volume).

Can it be a photonic process?

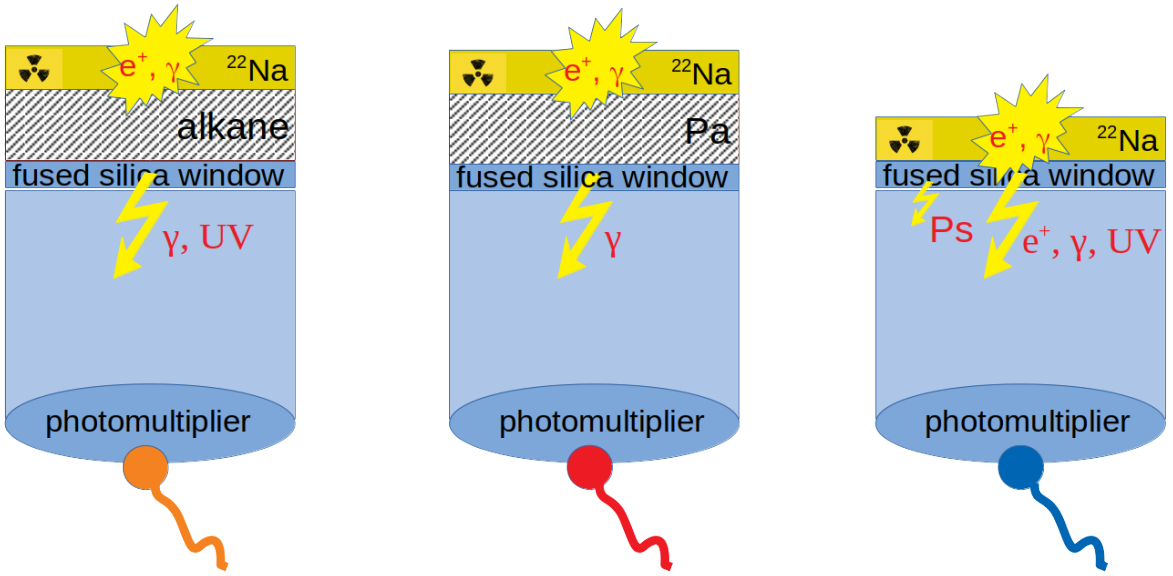
!!!New kind of spectroscopy??? $\Longleftarrow$



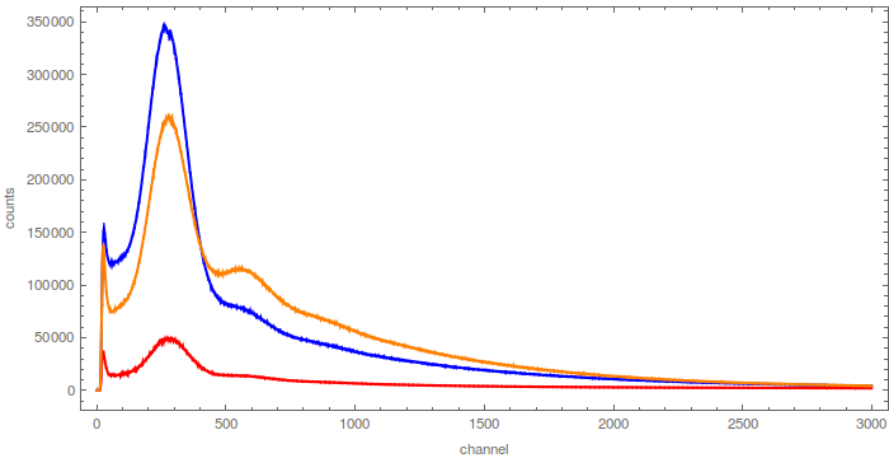


Violet light = 2.95–3.10 eV

N-UV is detectable by photomultiplier



Photomultiplier window transmittance: 200-600nm (6.2 – 2.1eV)



UV-Vis sources:

- Cherenkov radiat. (E_k >0.26MeV),
- ion recombinations (N-UV $\leftrightarrow$ F-UV),
- Ps formation (???).

MePS at HZDR

HZDR

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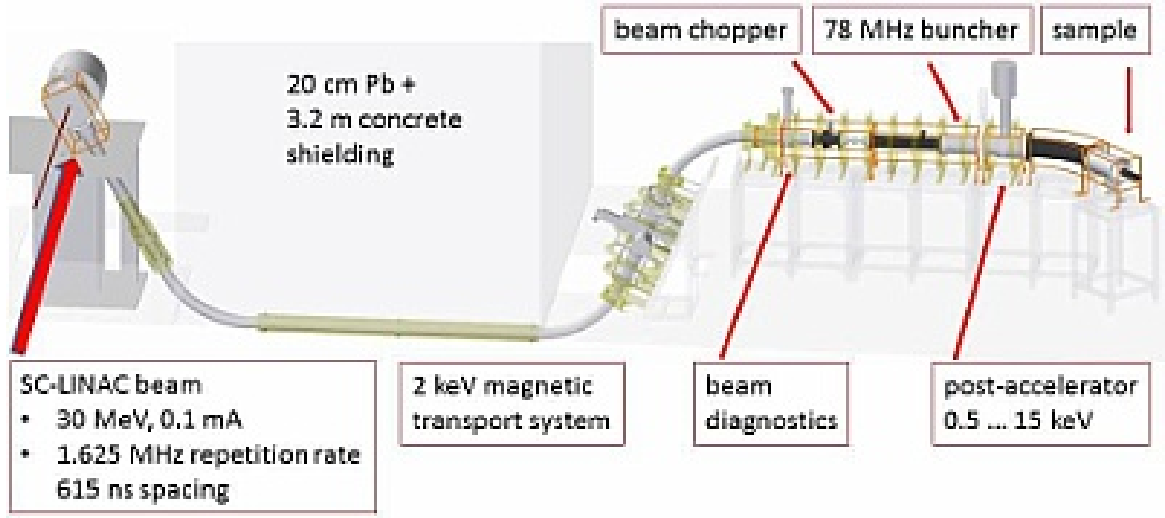
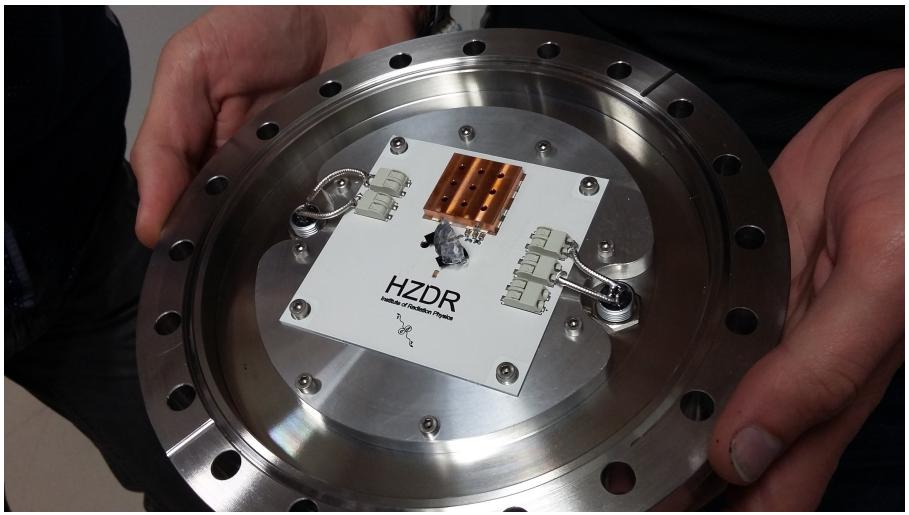
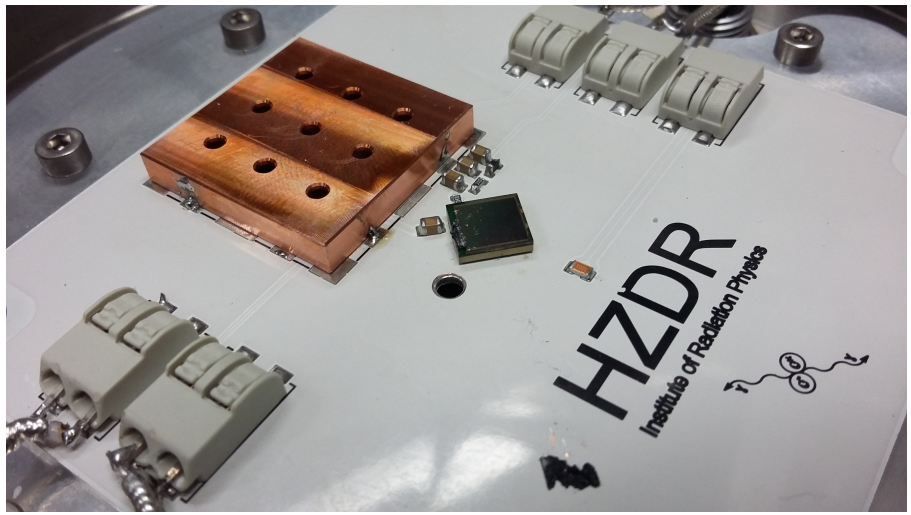


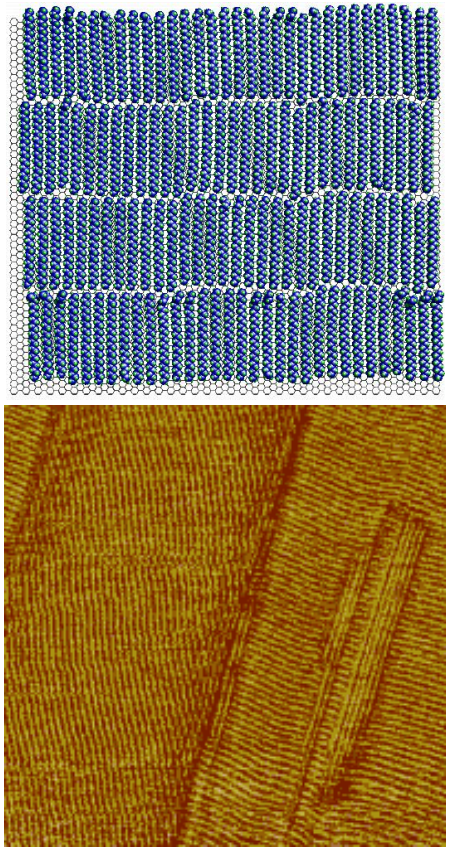




- The MEPS system at ELBE was used as a monoenergetic positron source. Positron beam energy: 2keV.
- Single photon detector: Hamamatsu S13360-6075PE photo sensor with 6400pix of a size 75μm cooled by a fluid. Detected wavelenght of quanta: 900nm-320nm (1.4eV-3.9eV).
- Temperature of a detector and the sample: -2°C.
- Acquiris DC282 10-bit digitizer (sample rate: 2 GSPS).
  Channel 1: CeBr analog signal.
  Channel 2: logical signal.
  Channel 3: SiPM analog signal.
  Statistics: 1.5M coincidence events for each sample.
- Samples: Alkane and porous silica samples fixed with double-sided carbon tape.

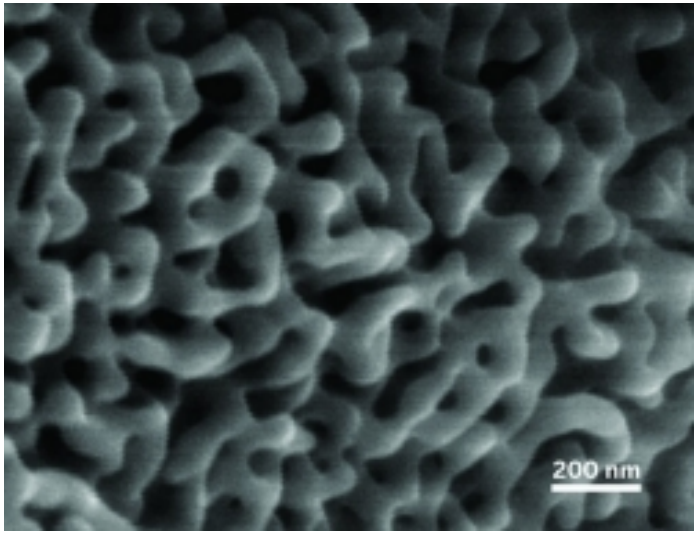
samples

n-alkane



goolge search

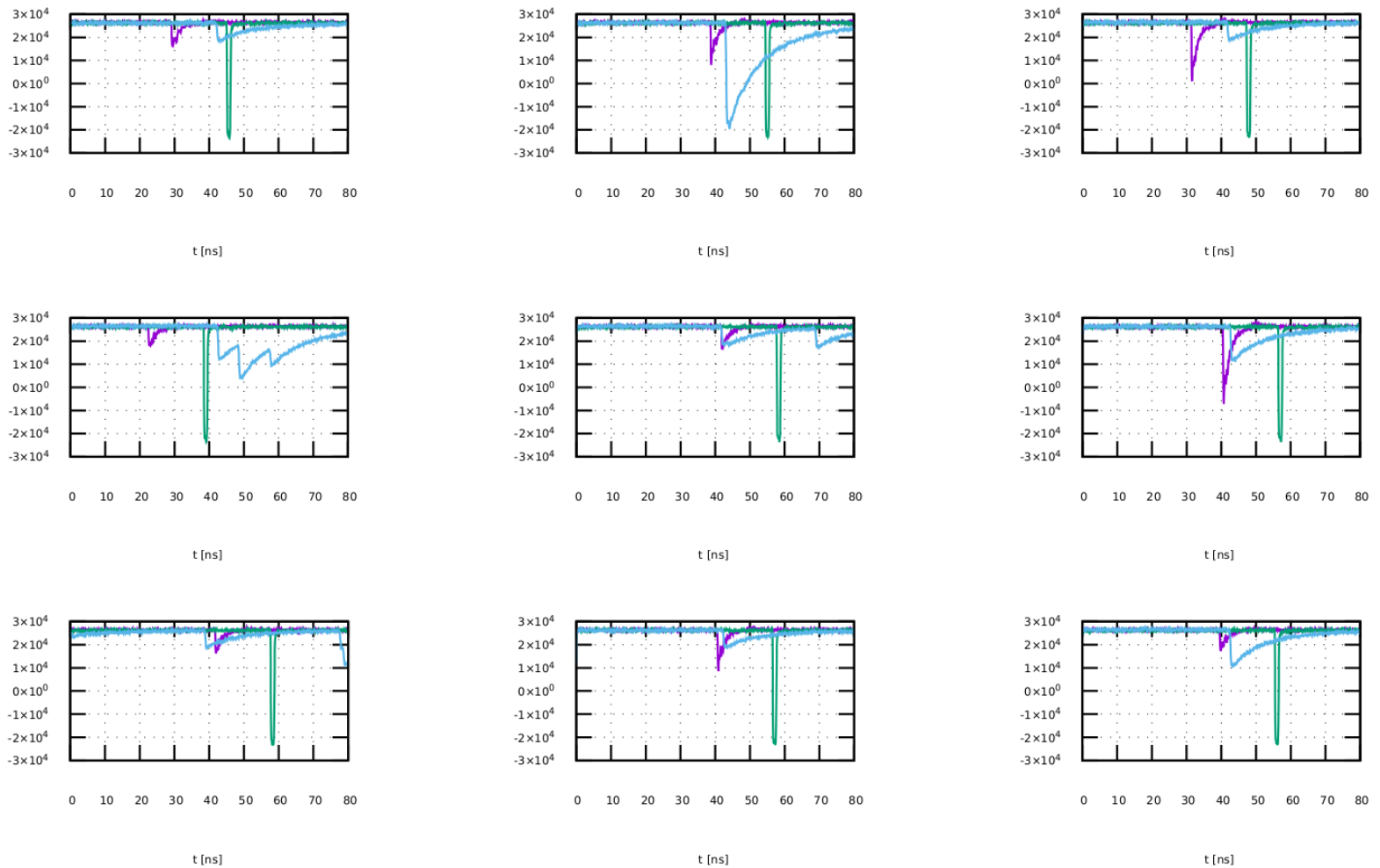
porous silica



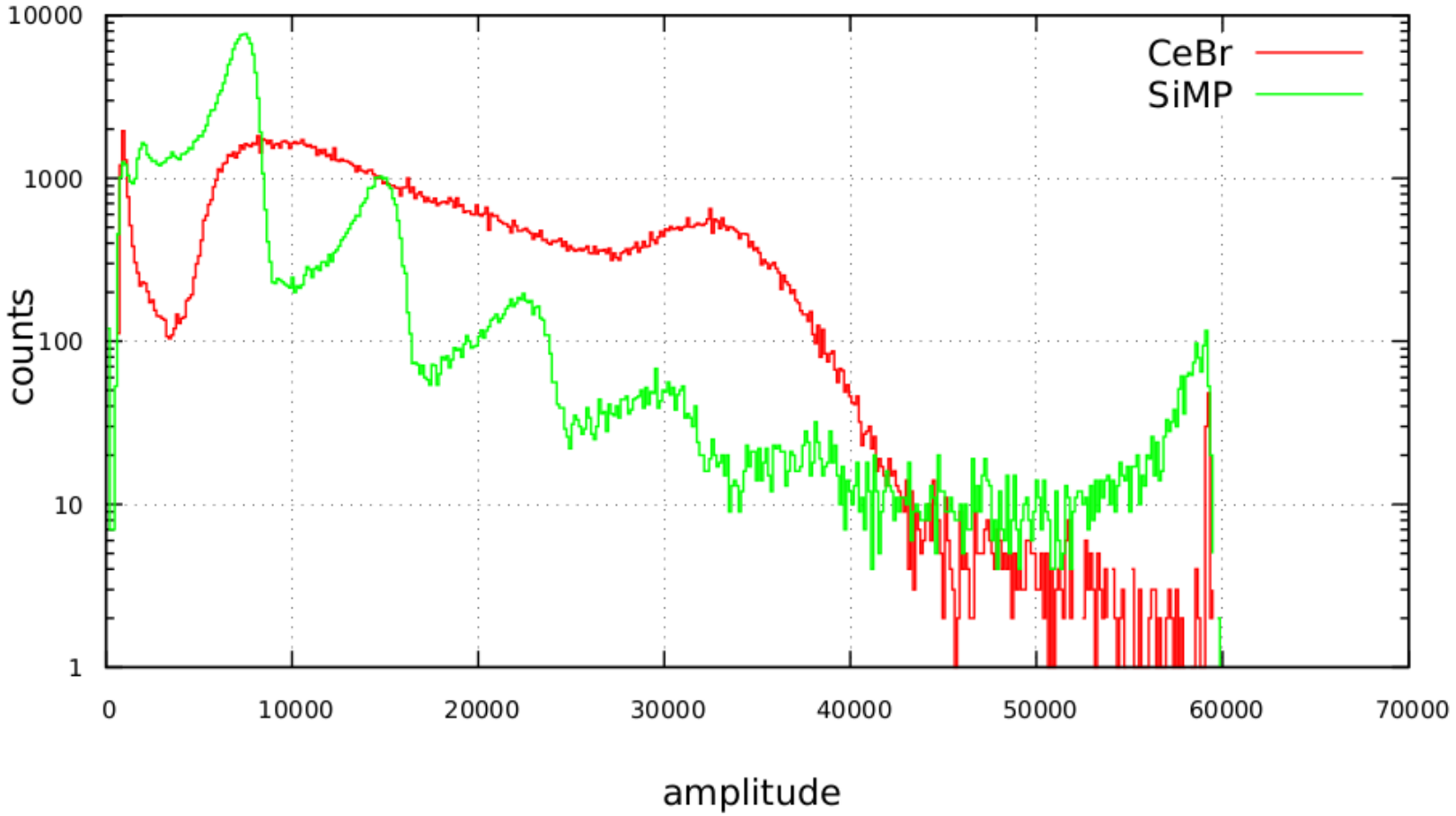
goolge search

timestamps – CeBr, SiPM, RF

It is known that low-energy positrons produce enhanced luminescence (compared to electrons) in some materials. Here, a considerable amount of photonic event are multi-peak signals.

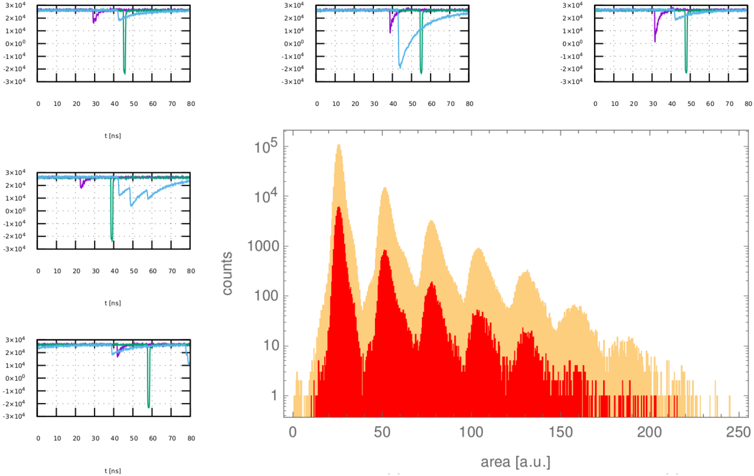


pulses' amplitude spectrum

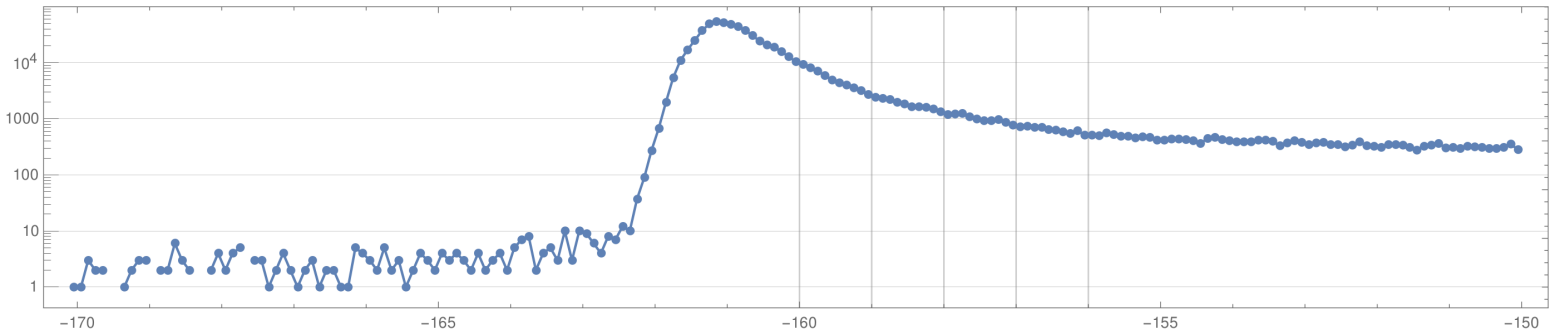


photonic pulses' area

The number of photons in a pulse is proportional to the area of them.



e^+ lifetime spectrum



Lifetime decomposition (comparison to PALS)

From Ps annihilating on the surface of the samle or outside the sample

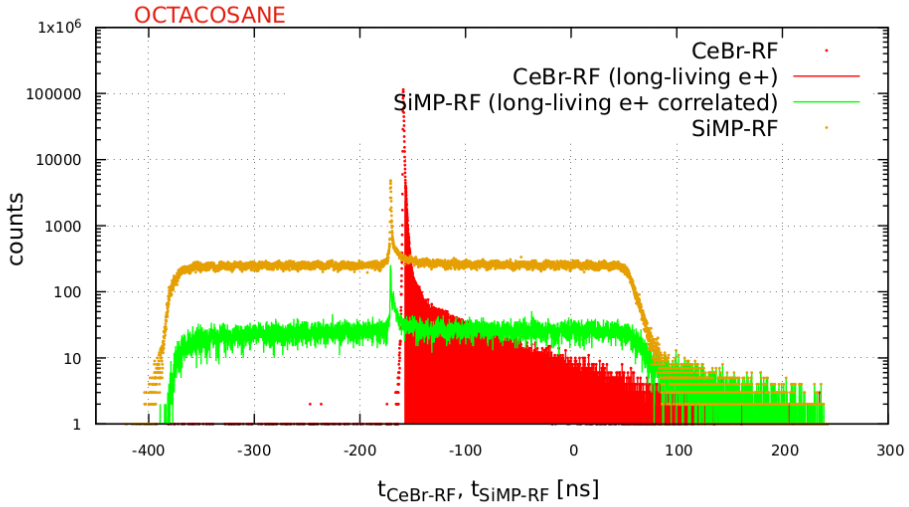
interlammellar o-Ps comp. known from PALS

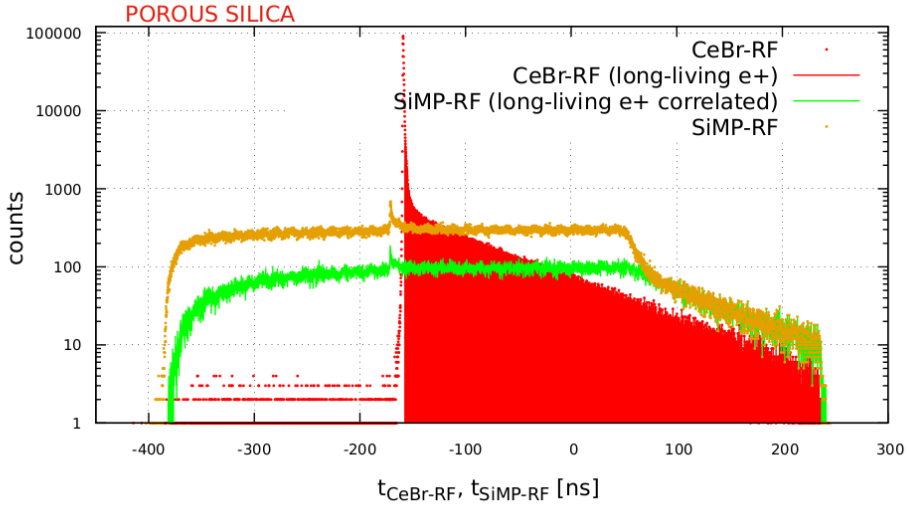
	I_1 [%]	τ_1 [ns]	I_2 [%]	τ_2 [ns]	I_3 [%]	τ_3 [ns]	I_4 [%]	τ_4 [ns]	I_5 [%]	τ_5 [ns]	I_6 [%]	τ_6 [ns]
octacosane	35.1	0.09	41.4	0.38	18.2	1.43	1.85	7.18	3.34	57.85	-	-
silica 51ns fixed	12.1	0.06	40.4	0.28	16.3	1.16	2.6	6.42	2.4	51.0	26.2	79.8

even 21% for PALS positrons!
(diffusion outside of the sample)

pore's o-Ps comp. known from PALS

e^+ – photon coincidences





fraction of Ps vs. fraction of their photons

E_t – Total number of the annihilation events (forming the positron lifetime spectrum) – red dots.
P_t – number of photons related to them – orange.
E_l – annihilation events from long-living positrons, i.e. t $>$ 2ns – solid red area.
P_l – corresponding photonic spectrum – green.

$$E \equiv \frac{E_l}{E_t}, \quad P \equiv \frac{P_l}{P_t}$$

$E < P$??? If yes, there are extra photons for Ps.
But our calculations show that ...

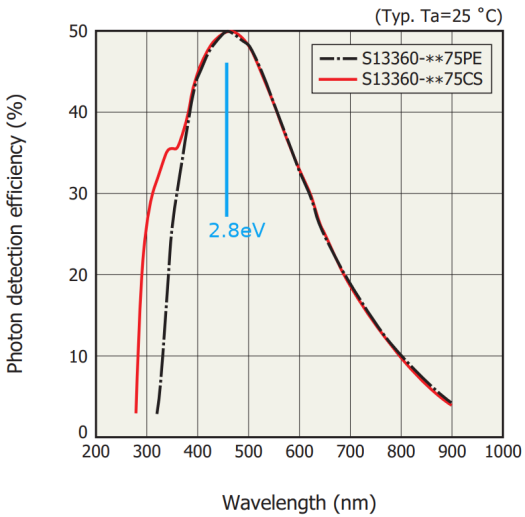
Conclusion

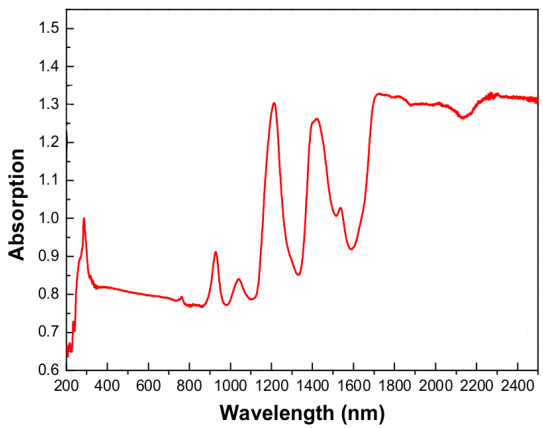
E=0.102, P=0.101 (octacosane)
E=0.324 and P=0.324 (porous silica)

Possibly, Ps creation does not produce extra photons ☹

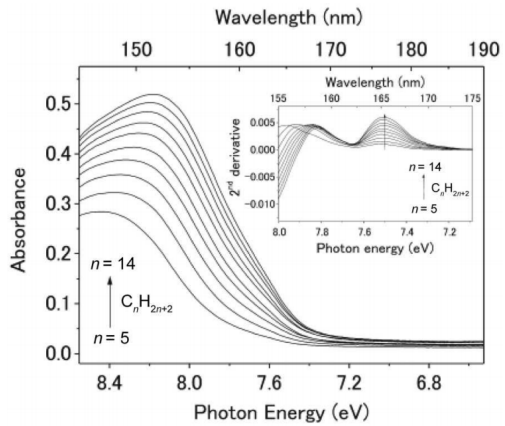
Possible issues – technical matter

- too small fraction of bulk Ps
- low efficiency of photon detection (SiPM efficiency, absorption)
- inadequate photon energy detected range
- too high temperature (phonons are possible too)



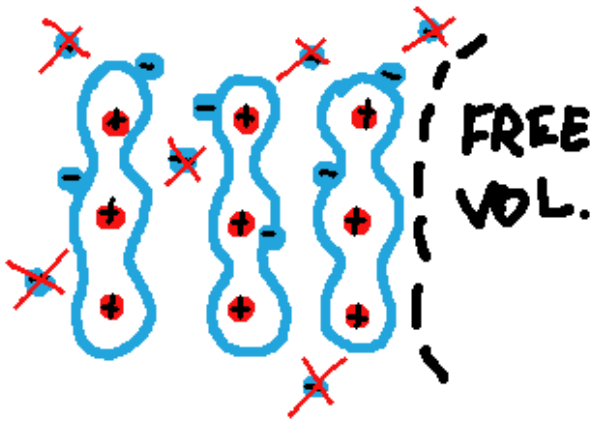


Y. Li, Y.A. Samad, *at al.*:
J. Mater. Chem. A, 2014, 2, 7759-7765



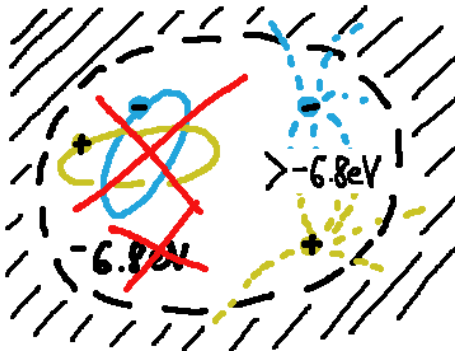
Y. Ozaki, Y. Morisawa, *at al.*:
Appl. Spectrosc., 66, 1-25.

Possible issues – theoretical concepts



Initial state: qusi-free $e^+ - e^-$ pair does not exist $\Rightarrow$ Ps formation with valence electrons require energy intake

Final state: Ps in a free volume cannot be identified with vacuum Ps for alkanes $\Rightarrow$ not so big energy gain during transition

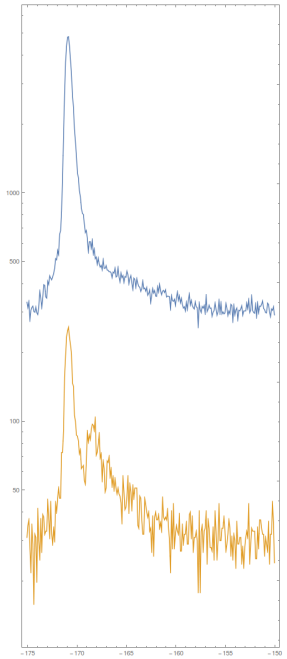


Way of transition: intermolecular transition of e^+ in alkanes require additional work $\Rightarrow$ more costly process of transition

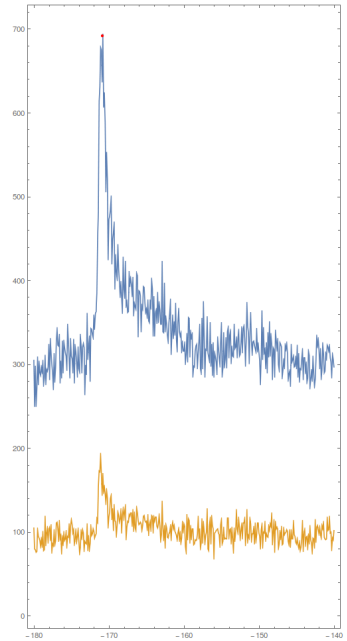
What to do next ...

We are going to repeat the experiment with modifications:

- to record more events
- to change the range of wavelenght detection
- to increase the energy of positrons (more bulk Ps)
- to decrease temperature (lesser energy dissipation)
- to apply a pressure (decrease distances = increase the e^+ intermolec. transitions)



Octacosane



Porous silica

□