

Nanophysics for scintillation registration: Advantages and limits

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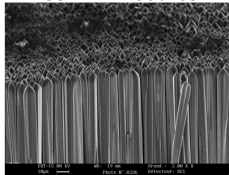
Shapping scintillators

Single Crystal



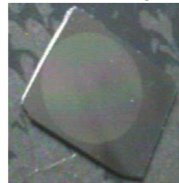
Many applications

CsI:Tl Needles



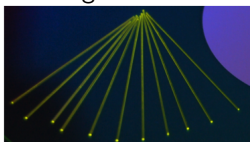
Medical x-ray imaging

Thin films



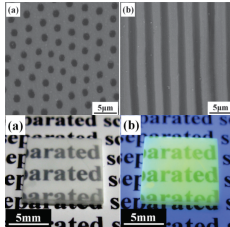
High resolution x-ray imaging

Inorganic Fibers



Calorimetry?

CsI-NaCl eutectic



@Canon, Adv. Mat. 2012

Phosphor powder



x-ray imaging

Shapping scintillators: Nano?

- New physics?
- New materials?
- New functionalities?



This is our job.

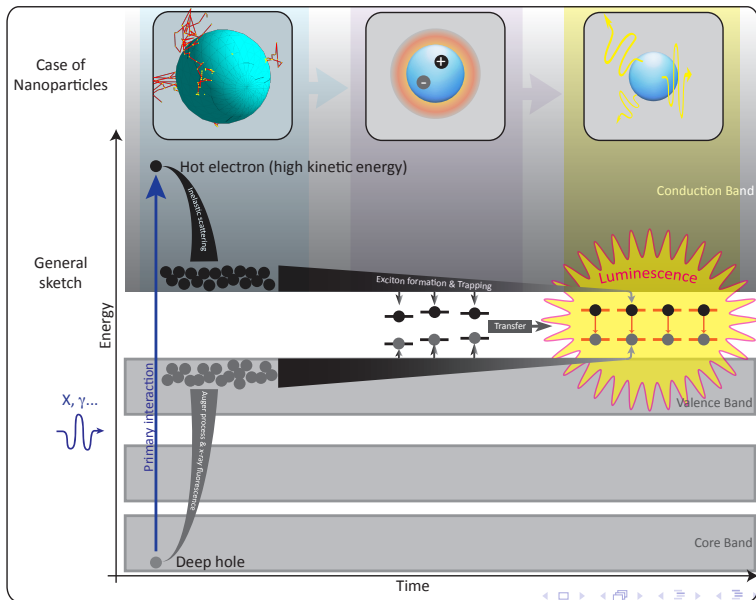
Nanoscintillators: Bibliography

Web of Sciences $\rightarrow \sim 130$ papers combining nano* x Scintillat*
(except nanosecond) (01/2013)

- 66 correspond to synthesis procedures of compositions of interest for scintillation
- 16 describe some scintillation properties
- 44 deal with some potential applications (most of them are about precursor for transparent ceramics)
- 4 are general/review papers

\rightarrow rather open field

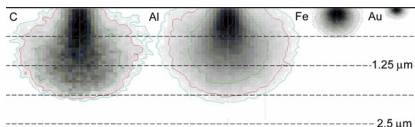
Processes: Specificity of nanosizes?



New Physics: β .S.Q (with A.L.Bulin & A.Vasil'ev)

Distribution of energy deposition after primary interaction

15keV electrons in various materials



<http://www4.nau.edu/microanalysis/Microprobe-SEM>

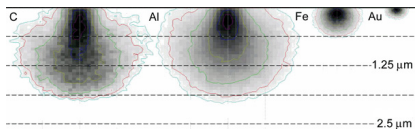
→ dimension \gg nanosizes

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Distribution of energy deposition after primary interaction

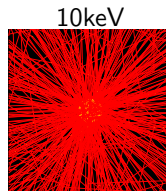
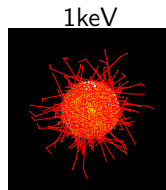
e^- created in a NP of 50nm
case of (Gd_2O_3)

15keV electrons in various materials



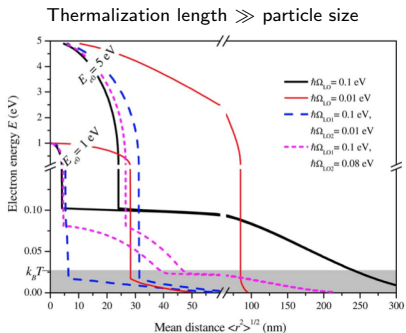
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→ dimension \gg nanosizes



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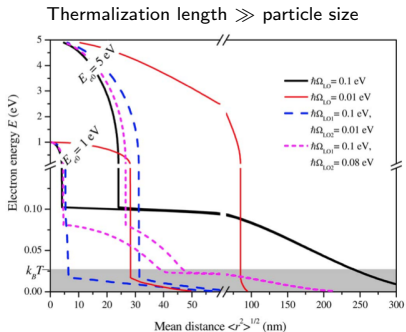
Interaction with Phonons



Kirkin et al. IEEE-TNS 2012

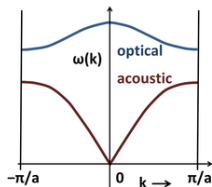
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Interaction with Phonons



Kirkin et al. IEEE-TNS 2012

Confinement on phonon energy?



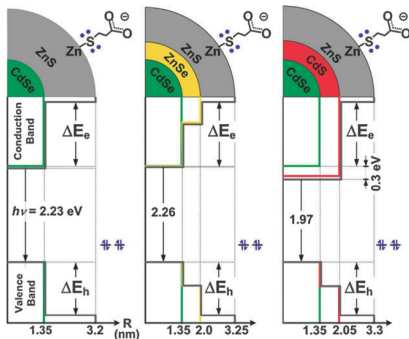
- Optical branch \rightarrow flat
- nano $\rightarrow \lambda_{max} \geq 2L_{nano} \rightarrow k \sim 0$ "vanish"
- no effects on optical modes
- cut off of acoustical mode

probably no effect of nano sizes regarding this aspect

New Physics: β .S.Q

Potential barrier for low energy electrons

Nanotechnologies use to play with potential barriers and Gap

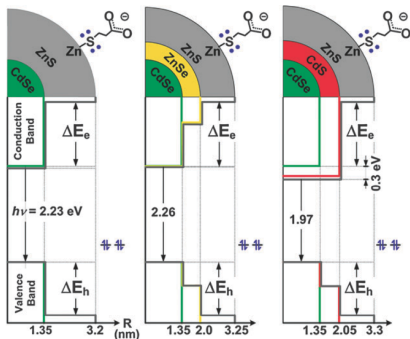


Bonghwan Chon, PhysChem.ChemPhys, 2009

New Physics: β .S.Q

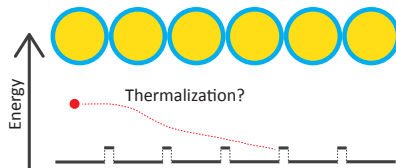
Potential barrier for low energy electrons

Nanotechnologies use to play with potential barriers and Gap



Bonghwan Chon, PhysChem.ChemPhys, 2009

Assembly of core-shell system might create structured potentials



Effect on spacial distribution of charges during relaxation \rightarrow non-proportionality?

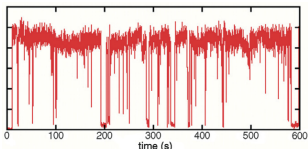
Excitons

- As for low energy electrons and holes, excitons have a mobility
 - effect on the rise time in case of transfer to activator?
 - potential barrier at the particle edge?

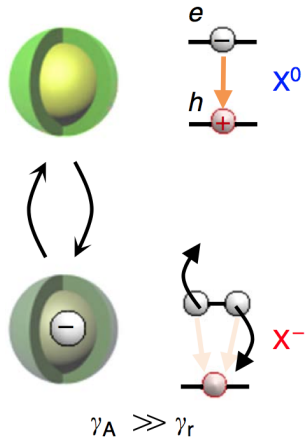
New Physics: β .S.Q

Excitons

- As for low energy electrons and holes, excitons have a mobility
 → effect on the rise time in case of transfer to activator?
 → potential barrier at the particle edge?
- Charging effect → Auger process instead of radiative recombination (blinking of QD)



Smider et. Al., Materials Today, 2001



Galland et. al., Nature Comm., 2012

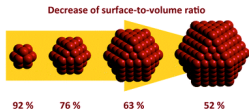
Traps

Traps might be different in nanoparticles

Stress



Large Surface/Volume ratio



P.Sonström et. al., Phys.Chem.Chem.Phys, 2011

Pt, 2nm \rightarrow 50% of the atoms at the surface

New Physics: β .S.Q

Traps

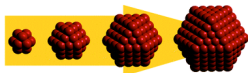
Traps might be different in nanoparticles

Stress



Large Surface/Volume ratio

Decrease of surface-to-volume ratio



92 %

76 %

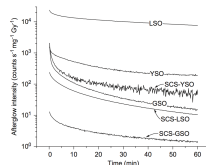
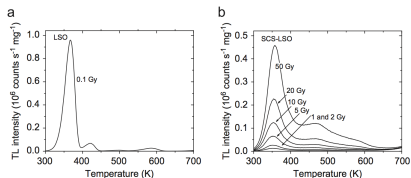
63 %

52 %

P.Sonström et. al., Phys.Chem.Chem.Phys, 2011

Pt, 2nm \rightarrow 50% of the atoms at the surface

Exemple with LSO (25-100nm) as precursor for ceramics



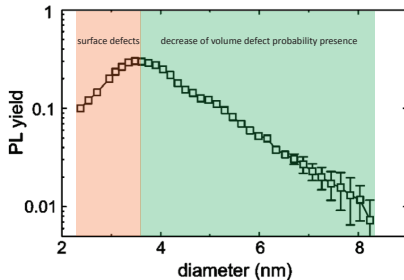
Yukihara, J.of.Lum, 2010

Effect on afterglow, Bright Burn / might be good for Photostimulation applications

Traps: example with Silicon

- Indirect band gap \rightarrow the yield is driven by the defect concentration
- at small sizes, the probability to find a bulk defect is smaller
- at very small sizes, surface defects \rightarrow requires surface passivation

Evolution of the Light Yield with the size

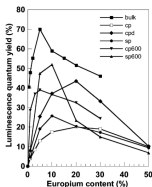


Ledoux, Applied Phys. Let., 2002

New Physics: β .S.Q

Surface = bad for luminescence

- Well-known in semi-conductor QD
→ confinement of charges
- in doped insulator, a lot of activators are next to the surface
→ surface treatment are required

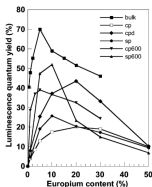


Huignard et. al., J.Phys.Chem. B, 2003

New Physics: β .S.Q

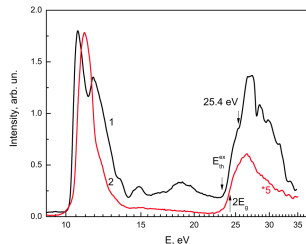
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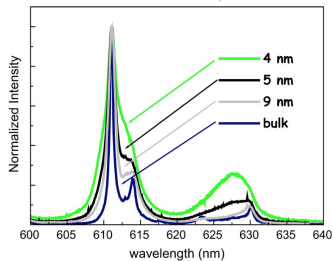
STE emission of 20nm and 140nm CaF_2 particles
→ effect of thermalization length



Vistovsky et. al., J. of Applied Phys., 2012

- no improvement and even some losses are expected in very efficient materials
- but potential improvements for materials having non-radiative recombinations

Effects on Luminescence

Disorder \rightarrow BroadeningCase of nano- $Gd_2O_3 : Eu^{3+}$ 

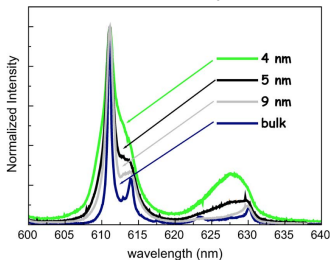
Dujardin et. al., IEEE TNS, 2010

New Physics: $\beta.S.Q$

Effects on Luminescence

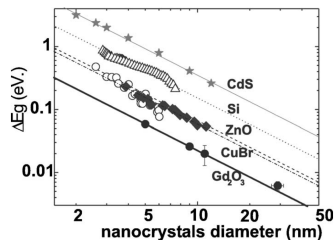
Disorder \rightarrow Broadening

Case of nano- $Gd_2O_3 : Eu^{3+}$



Dujardin et. al., IEEE TNS, 2010

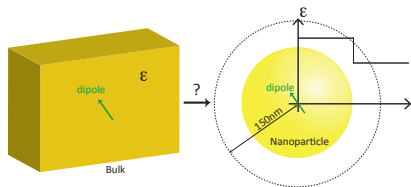
Ionic materials \sim No confinement effect on gap



Mercier et. al., J.Chem. Phys., 2007

Effects on Luminescence: Dielectric confinement

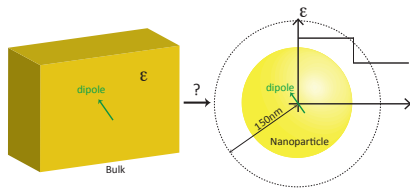
The radiative decay time depends on the index of refraction which is a **macroscopic** parameter



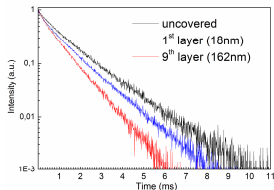
New Physics: $\beta.S.Q$

Effects on Luminescence: Dielectric confinement

The radiative decay time depends on the index of refraction which is a **macroscopic** parameter



several models: Fully macroscopic, empty and virtual cavity



LeBihan et. al., PRB, 2008

→ nano-LuAG:Ce $\tau = 117\text{ns}$ vs 58ns in bulk
(Barta et.al. J.Mat.Chem,2012)

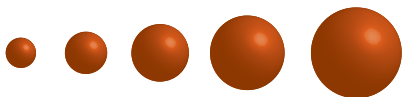
In addition, competition between radiative and non-radiative

probabilities:
$$\eta = \frac{W_{rad}(n)}{W_{rad}(n) + W_{nonrad}}$$

Difficulties

Experimental difficulties

The physicist wants

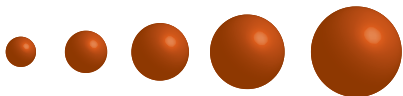


- size gradient over a wide range
- sharp size and shape distribution
- identical surface states
- single phase domain
- ...

Difficulties

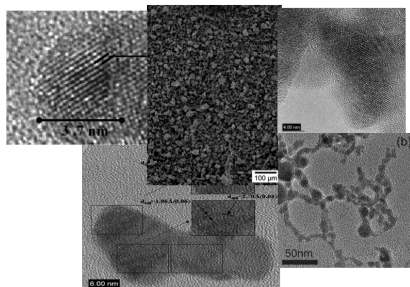
Experimental difficulties

The physicist wants



- size gradient over a wide range
- sharp size and shape distribution
- identical surface states
- single phase domain
- ...

The chemist provides



→ difficult for comparison

Applications

nanoparticles are small

→ they can be mixed into matrix while preserving transparency

ZnS:Mn NP dispersed in PMMA



<http://chm.tu-dresden.de>

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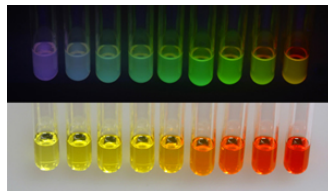


<http://chm.tu-dresden.de>

Colloïdale solution



©ILM



<http://www.webexhibits.org>

→ hybrides & liquid scintillators

Applications-Hybrides

Doping with nano particles

Claimed advantages: low cost as compared to single crystals

CdSe/ZnS Qdots in glasses

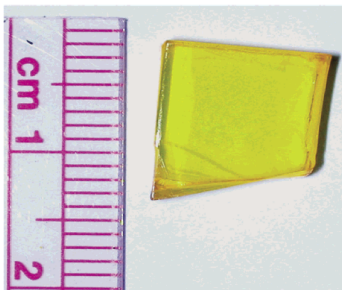
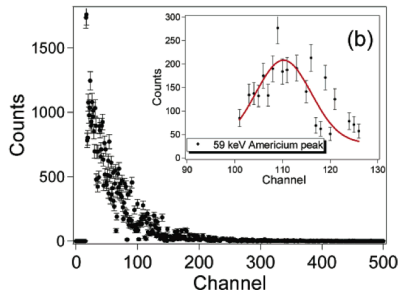


Figure 1. Picture of a 1/16 in. thick nanoporous glass slab impregnated with CdSe/ZnS quantum dots emitting at 510 nm.

Letant et. al. NanoLetters, 2006

Photopeak?



Nevertheless, some scintillation can be observed

Applications-Hybrides

In the same spirit, but with $LaF_3 : Ce^{3+}$ but in organics

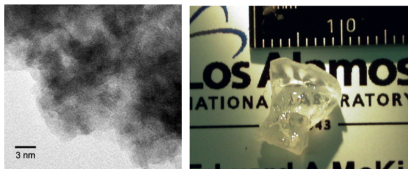
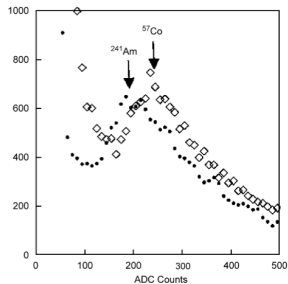


Figure 1. Transmission electron microscopy image of the $Ce:LaF_3$ particles (left) and a 1cm thick oleic-based nanocomposite of $Ce:LaF_3$ with nearly 30% volume loading of the phosphor.



McKigney et. al., NIMA, 2007

issue → high loading while preserving the transparency & properties

personal opinion :

not sure that it can compete with crystals regarding performances.

Application-Hybride

US Congres report



Detection of Nuclear Weapons and Materials: Science, Technologies, Observations

Jonathan Medalia

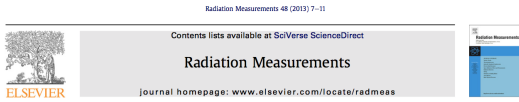
Specialist in Nuclear Weapons Policy

June 4, 2010

...However, unexpected nanoscale physics could impair energy resolution...

Application-Hybride

Doping organics with optically "active" nano particles specific functionality: neutron detection/ increase of density



GdBr₃:Ce in glass matrix as nuclear spectroscopy detector

Z.T. Kang, R. Rosson*, B. Barta, C. Han, J.H. Nadler, M. Dorn, B. Wagner, B. Kahn

Georgia Tech Research Institute, Georgia Institute of Technology, 525 Dalney St NW, Atlanta, GA 30332, USA



Fig. 1. Cerium-activated gadolinium bromide glass-matrix cylinder, 2.5-cm dia. × 3.0-cm height, under 365 nm UV illumination.

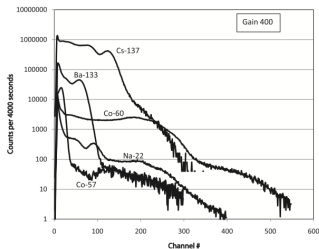


Fig. 3. Five gamma-ray spectra with 2.5-cm dia., 3.0-cm height, alumina-silica glass-matrix detector at gain 400.

Doping organics with optically "passive" nano particles

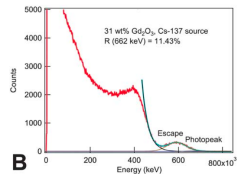
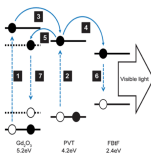
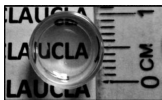
Journal of
Materials Chemistry C

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PAPER

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Synthesis of bulk-size transparent gadolinium oxide-polymer nanocomposites for gamma ray spectroscopy

Cite this: *J. Mater. Chem. C*, 2013, 1, 1970Wen Cai,^{1,2d} Qi Chen,^{1,a} Nerine Cherepy,³ Alex Dooraghi,² David Kishpaugh,³ Arion Chatzioannou,² Stephen Payne,³ Weidong Xiang² and Qibing Pei^{1a}

Application-New Materials

Improving Organic Scintillators

Journal of
Materials Chemistry C

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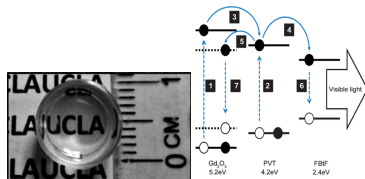
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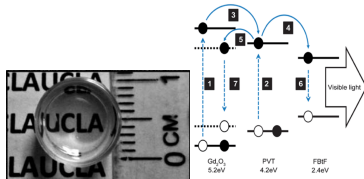
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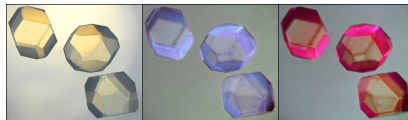
Out of traditional scintillators: MOF Metal Organic Framework

www.advmat.de

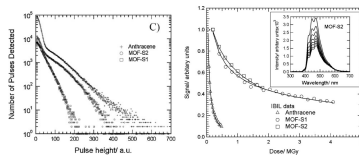
ADVANCED
MATERIALS

Scintillating Metal-Organic Frameworks: A New Class of Radiation Detection Materials

By F. P. Doty,* C. A. Bauer, A. J. Skulan, P. G. Grant, and M. D. Allendorf*



<https://share.sandia.gov>



Application-Liquid Scintillation

Various applications: metrology,
neutrino

Toluene + PPO molecule



- molecule \leftrightarrow active nanoparticles?
- Advantage: large Stokes shift
- Drawbacks: stabilization of high concentration & knowledge about energy transfers?

Application-Liquid Scintillation

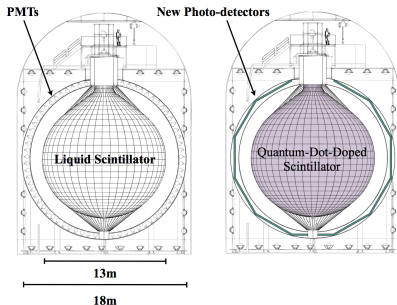
Various applications: metrology,
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Toluene + PPO molecule



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One example



Winslow et. al., JINST, 2012

In this case the Stokes shift is not so large,
cost???

Conclusion

new physics?

- yes: around the spacial distribution of excitation during relaxation
- yes: around the interactions with their surrounding (hybrid)

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- yes: optimum sizes are different from other field
- yes: can be combined with other compounds

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- yes: hybrides & nano composites
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worth investigating



