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Development of liquid scintillator and its application to Neutrino Physics

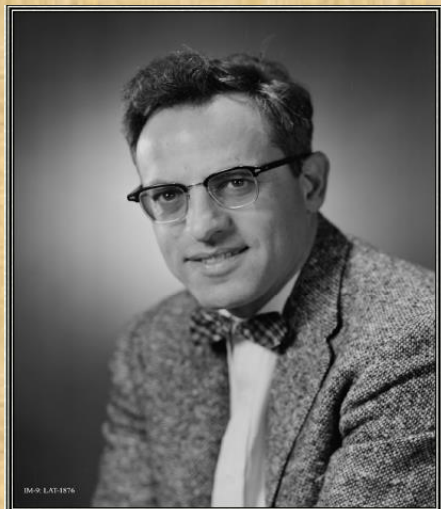
186th committee on radiation science and its application

FY2012 5th meeting

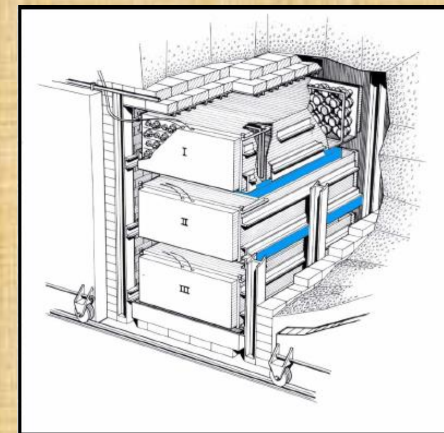
March 2nd , 2013

Yoshiyuki FUKUDA (Miyagi University of Education)

In the beginning of neutrino physics



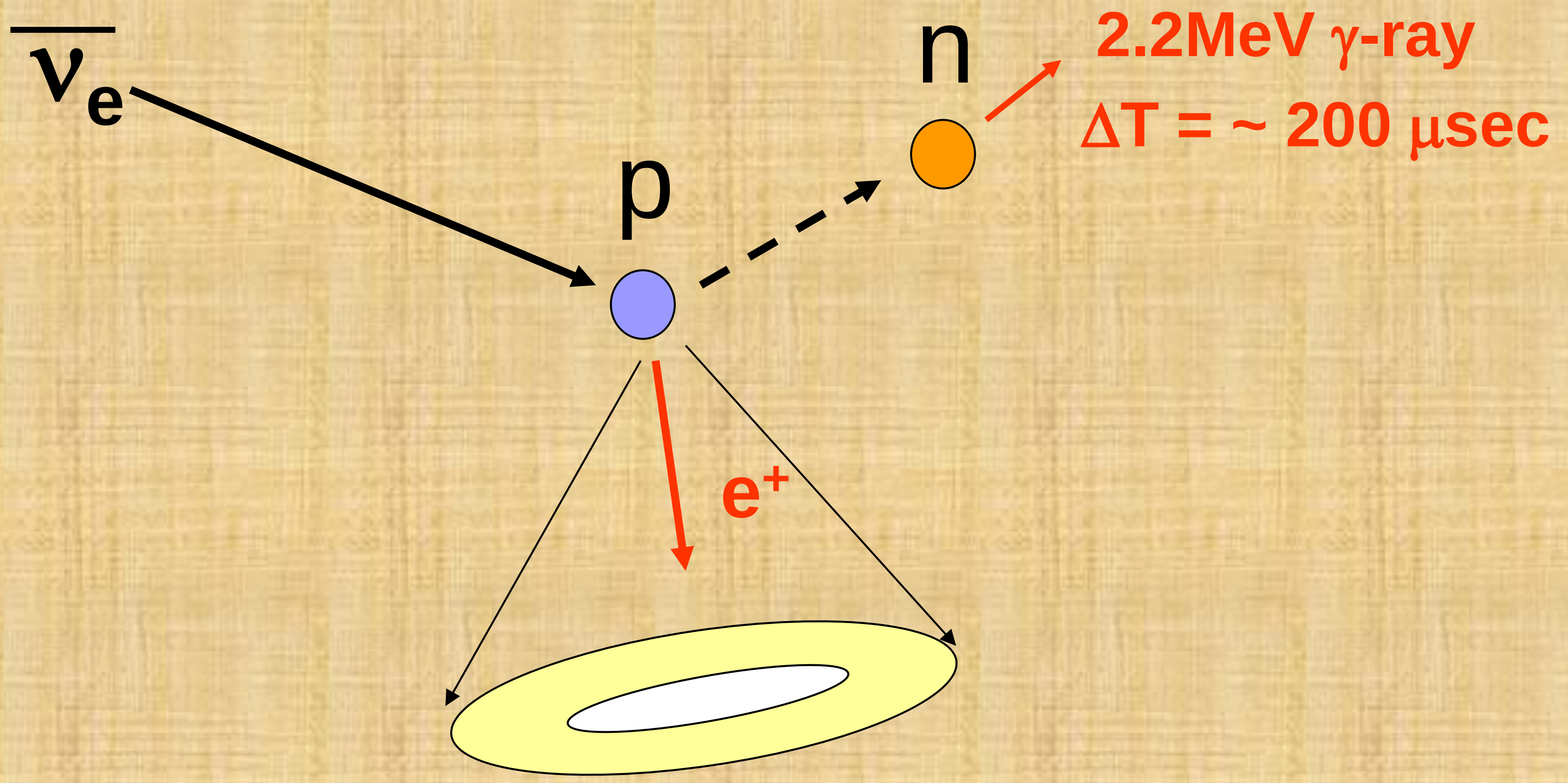
Frederick Reines



…with Clyde Cowan, another Los Alamos staff member. We initially considered the use of a nuclear bomb test as the source of neutrinos, but soon decided that the reactor at Hanford, Washington, would be better.

In 1956, it was succeeded to detect $\overline{\nu}_e$ using liquid scintillator from commercial reactor.

Detection method for anti-electron neutrino



No pre-activity

Very tiny post-activity with ΔT=~200μsec.
(trig. eff.=~1%)

Elementary particle

電荷

0

-1

-1/3

+2/3

軽い ⟷ 重い

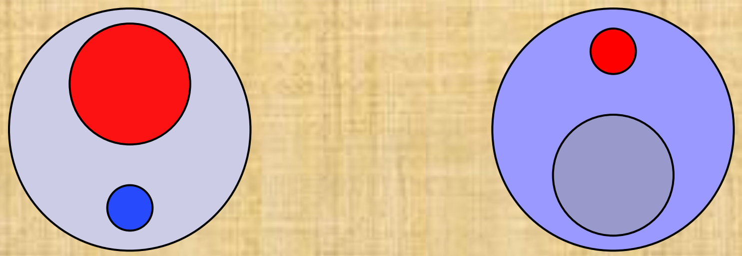
ニュートン1998年7月号

Neutrino oscillation

mass eigenstate and

ν_2(mass=m_2) ν_3(mass=m_3)

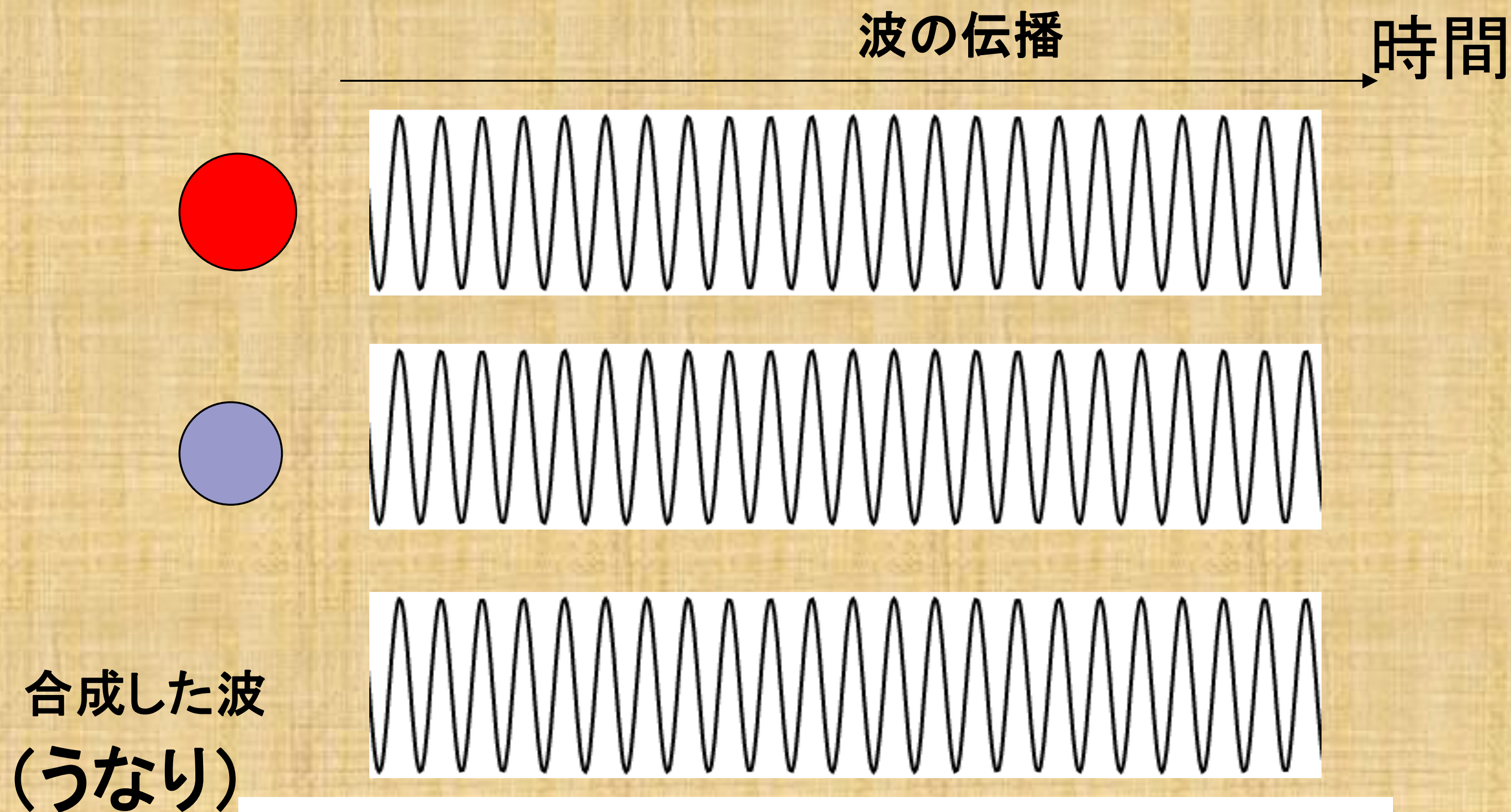
mixture of mass eigenstate ν2 and ν3



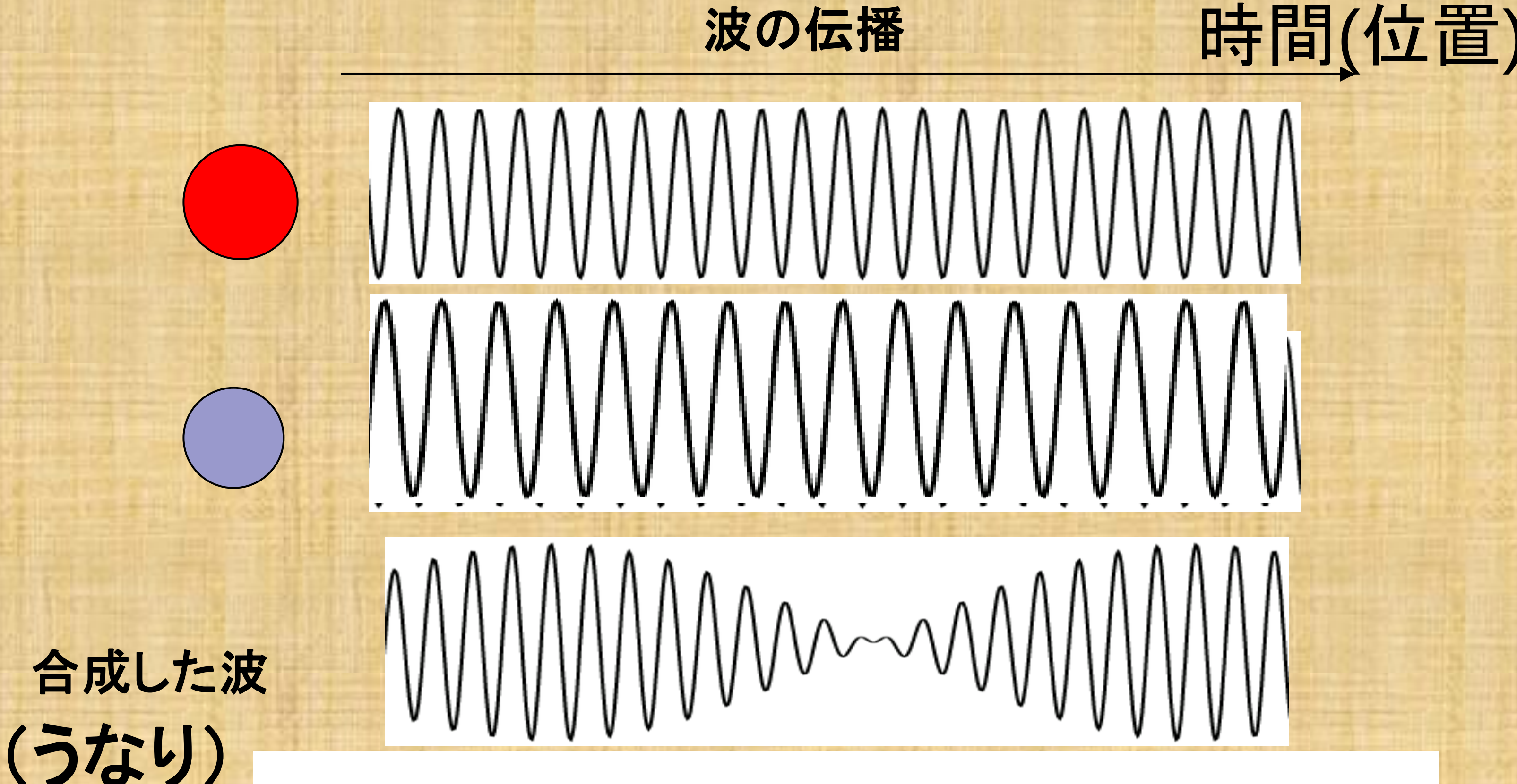
Muon neutrino Tau neutrino

Neutrino oscillation is easily explained by interference of wave.

If and are same mass, wavelength is same



if and are not same, wavelength is different



ee
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Neutrino oscillation experiments

If neutrinos have a finite mass, they will change flavor state in the pass of flight.

$$P(\nu_\alpha \rightarrow \nu_\beta) = \sin^2 2\theta \ \sin 2(1.27 \Delta m^2 L/E)$$

cf. assuming E=1GeV L=250km

$\Delta m^2 = 0.001 eV^2$ at maximum

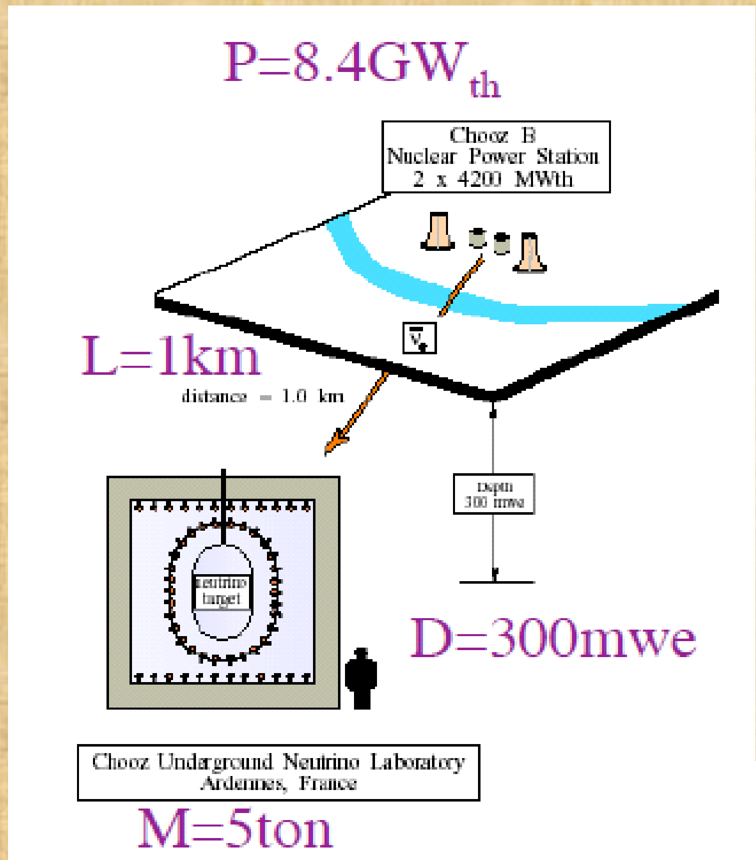
- Appearance experiment

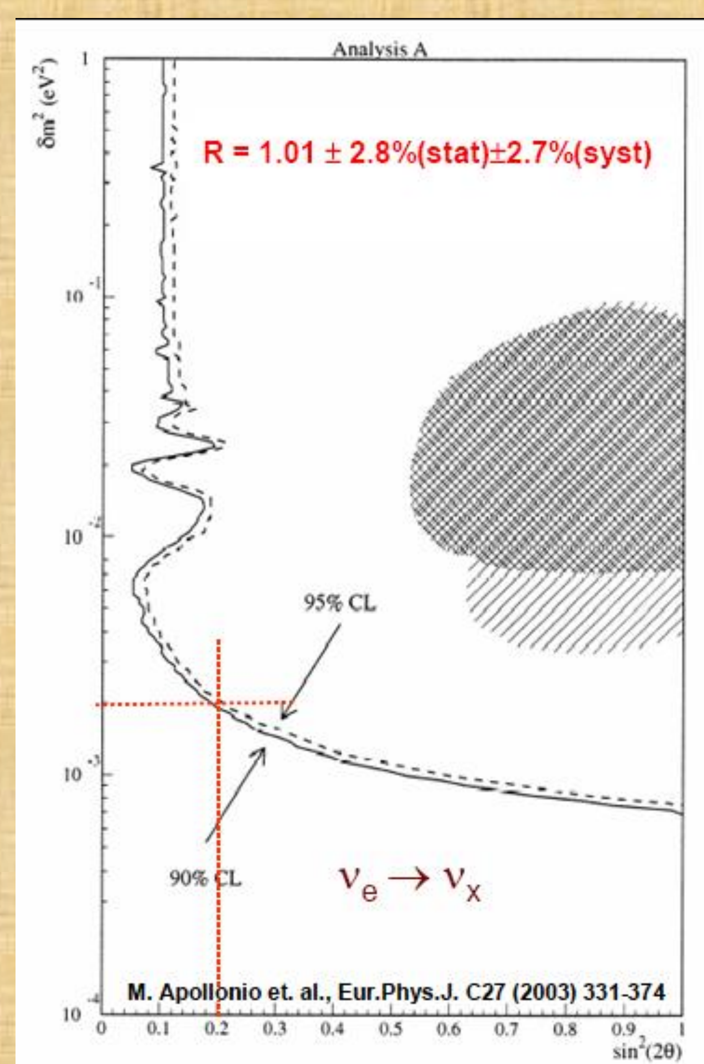
  Searching for neutrinos which does not exist at source

- Disappearance experiment

  Measurement of intensity of known flavor of neutrinos from source

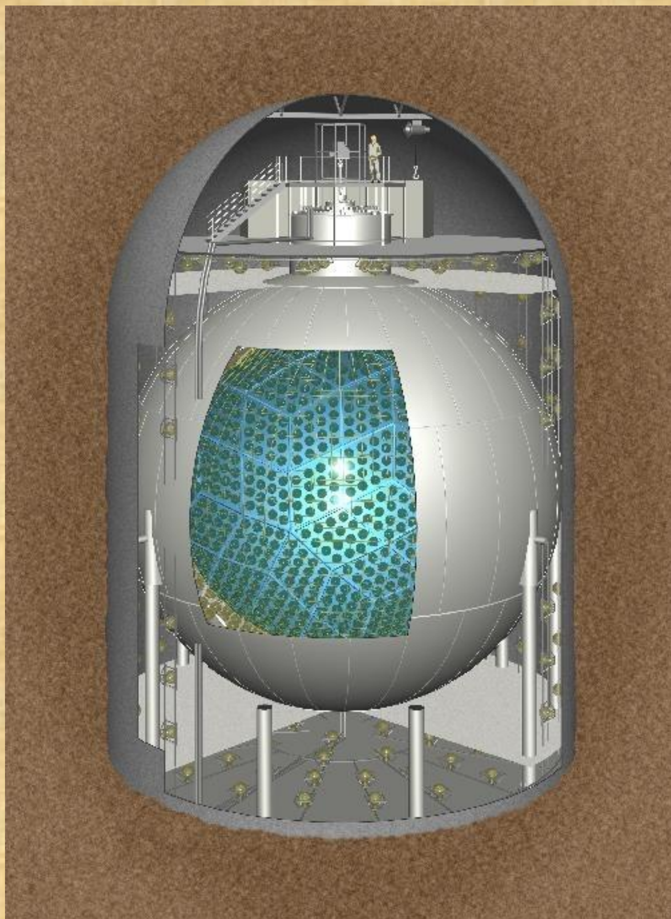
CHOOZ experiment



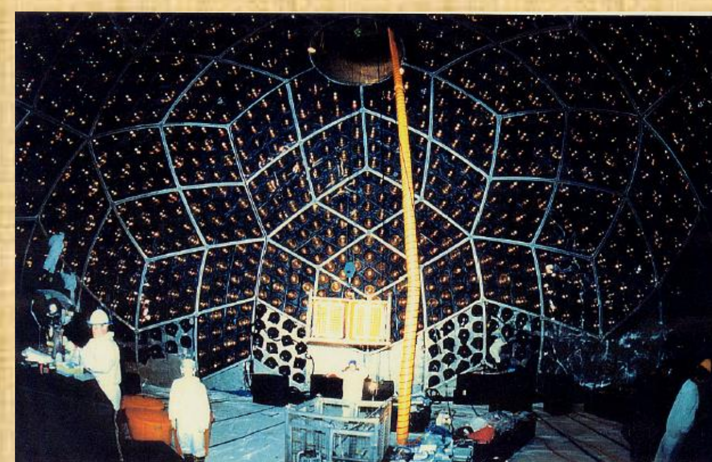


$$\sin^2 2\theta_{13} < 0.2 \ @ \ \Delta m^2_{13} = 2 \times 10^{-3} \mathrm{eV}^2$$

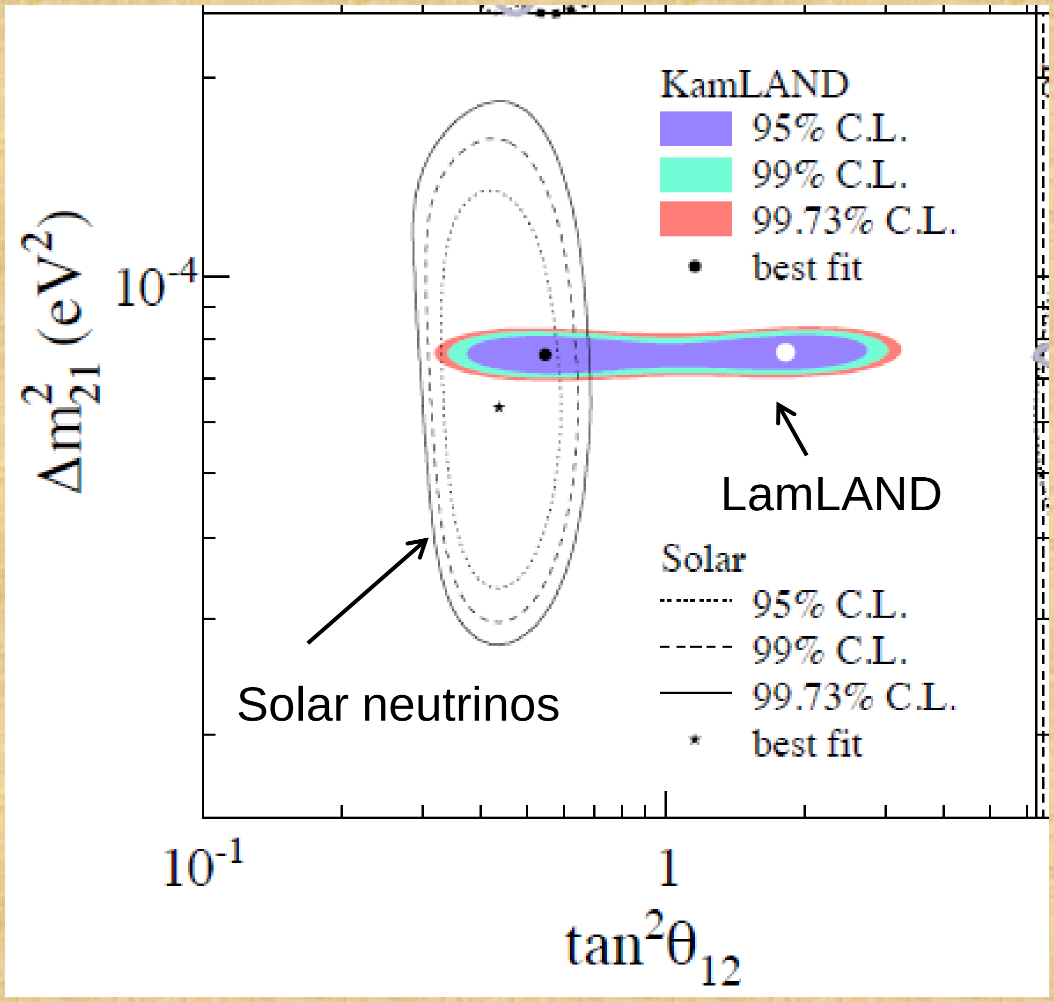
KamLAND experiment



Kashiwazaki –kariwa
Nuclear power station



Allowed region of oscillation parameters



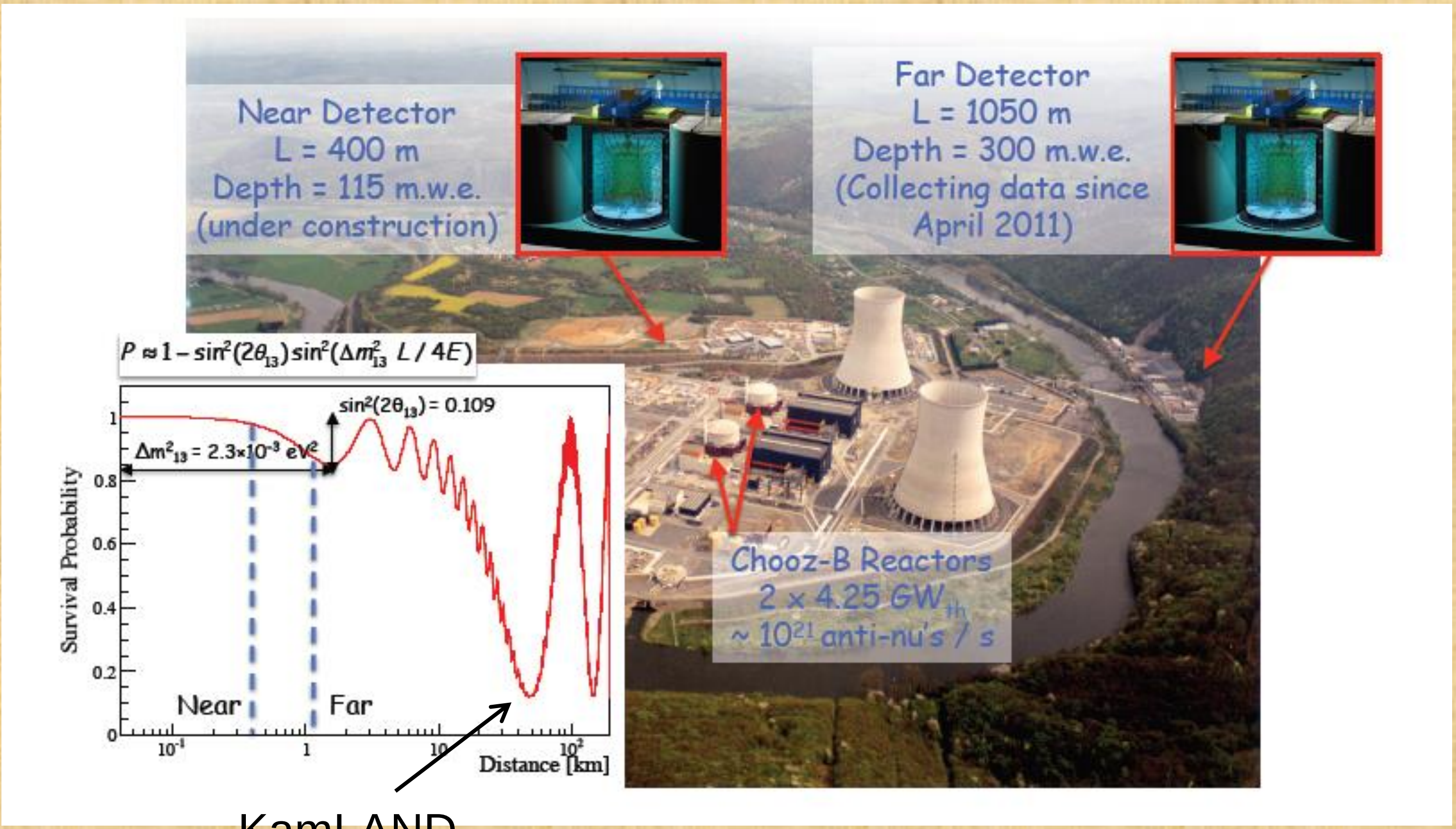
Consistent of results from solar neutrino experiments

Evidence of solar neutrino oscillation

Recent reactor experiments

$P(\nu_e \rightarrow \nu_e) = 1 - \sin^2 2\theta_{13} \sin^2(\Delta m^2_{13} L/4E) + O(10^{-3})$ (as Δm_{12}~solarν)

cf. E=4MeV and $\sin^2 2\theta_{13} = 0.1$



KamLAND

Discovery of finite θ_{13}

In 2012, the question of whether or not $\theta_{13} \neq 0$ had finally been answered by three reactor experiments…

Past

Nov. 2011 → Double Chooz: $\sin^2(2\theta_{13}) = 0.086 \pm 0.051$

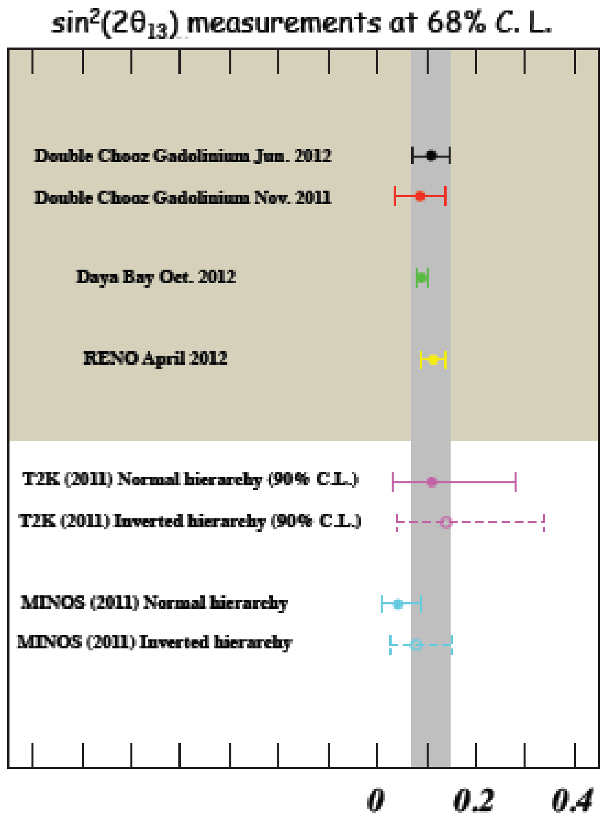
March 2012 → Daya Bay: $\sin^2(2\theta_{13}) = 0.092 \pm 0.017$

April 2012 → RENO: $\sin^2(2\theta_{13}) = 0.113 \pm 0.023$

June 2012 → Double Chooz: $\sin^2(2\theta_{13}) = 0.109 \pm 0.039$

Oct. 2012 → Daya Bay: $\sin^2(2\theta_{13}) = 0.089 \pm 0.011$

Present



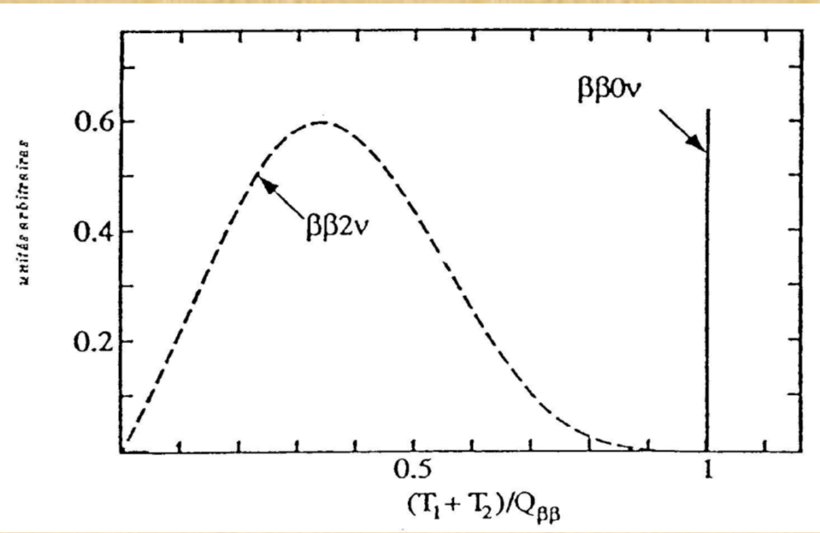
For the next couple years:

More precise θ_{13} = Improved search for CP-violation, mass hierarchy…

Neutrinoless double beta decay

$\beta\beta$ emitters with $Q_{\beta\beta}$ >2 Mev

Transition	$Q_{\beta\beta}$ (keV)	Abundance (%) ($^{232}Th = 100$)
$^{110}Pd \rightarrow ^{110}Cd$	2013	12
$^{76}Ge \rightarrow ^{76}Se$	2040	8
$^{124}Sn \rightarrow ^{124}Te$	2288	6
$^{136}Xe \rightarrow ^{136}Ba$	2479	9
$^{130}Te \rightarrow ^{130}Xe$	2533	34
$^{116}Cd \rightarrow ^{116}Sn$	2802	7
$^{82}Se \rightarrow ^{82}Kr$	2995	9
$^{100}Mo \rightarrow ^{100}Ru$	3034	10
$^{96}Zr \rightarrow ^{96}Mo$	3350	3
$^{150}Nd \rightarrow ^{150}Sm$	3667	6
$^{48}Ca \rightarrow ^{48}Ti$	4271	0.2



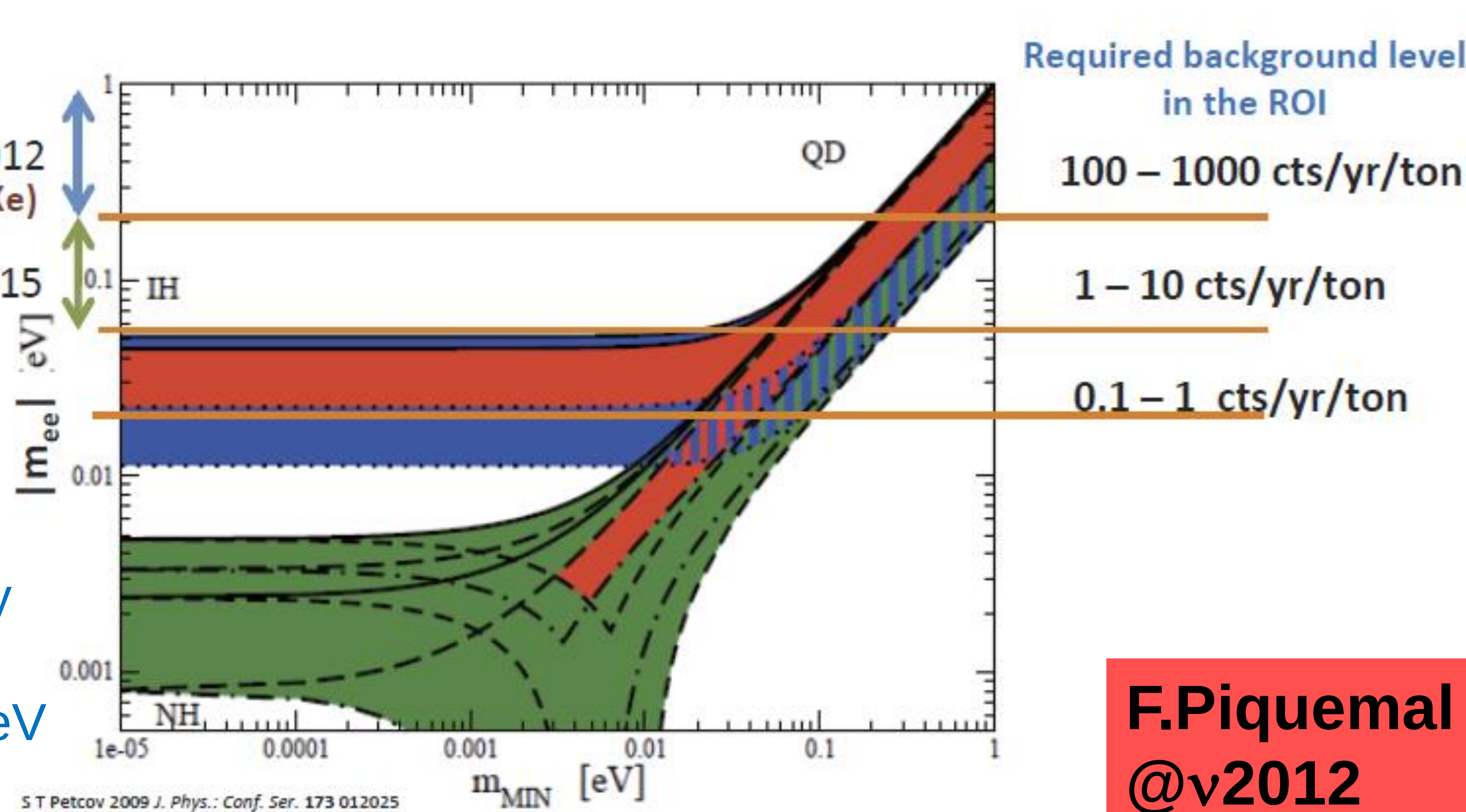
$$[T_{1/2}^{0\nu}(0^{+} \rightarrow 0^{+})]^{-1} = G_{0\nu}(E_0,Z)|M_{0\nu}|^2 \langle m_\nu \rangle^2$$

$T_{1/2}$~$a(Mt/\Delta EB)$ a: abundance M: mass

t: meas.time ΔE: energy res. B: BG rate

Requirement : Low BG, Large target mass, High energy resolution

For future experiments



high energy resolution 4%@2.5MeV

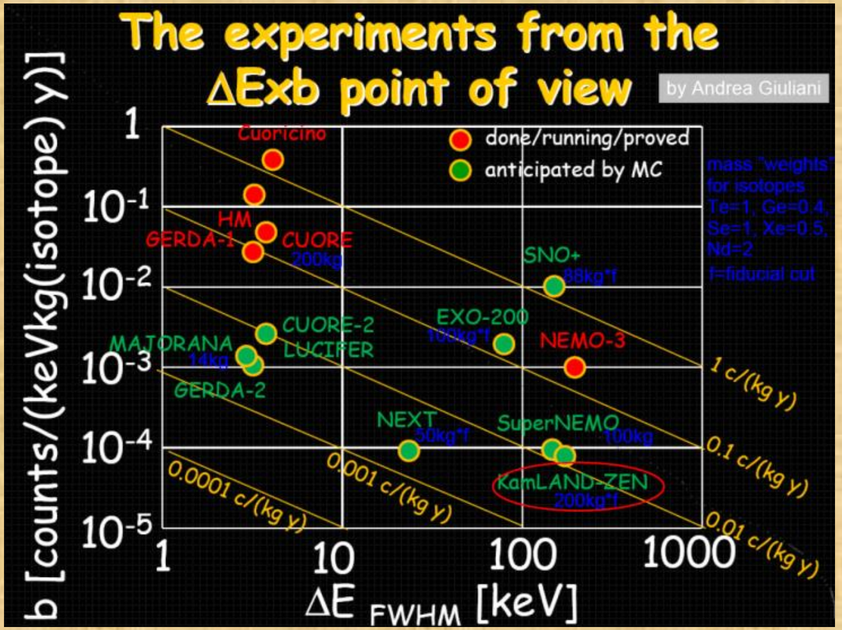
F.Piquemal @ν2012

http://kds.kek.jp/getFile.py/access?contribId=37&sessionId=16&resId=2&materialId=slides&confId=9151

~tons of target will be needed for next generation detector

Future experiments for Neutrinoless double beta decay

- Experimental requirement for BG rate and ΔE



- To achive m_ν < 100meV
  - high energy resolution 4%@2.5MeV
  - low background rate 0.01count kg^{-1} y^{-1}
  - ton scale of target

Liq. Scintillator is easy to scale up target volume

Studied isotopes

Piquemal@ν2012

^{116}Cd	COBRA $CdWO_4$
^{76}Ge	GERDA MAJORANA
^{82}Se	SuperNEMO LUCIFER
^{136}Xe	KamLAND-Zen EXO NEXT
^{100}Mo	$ZnMoO_4$ AMoRE
^{130}Te	CUORE
Nat(48)Ca	CANDLES
Nat(150)Nd	SNO+ Borexino

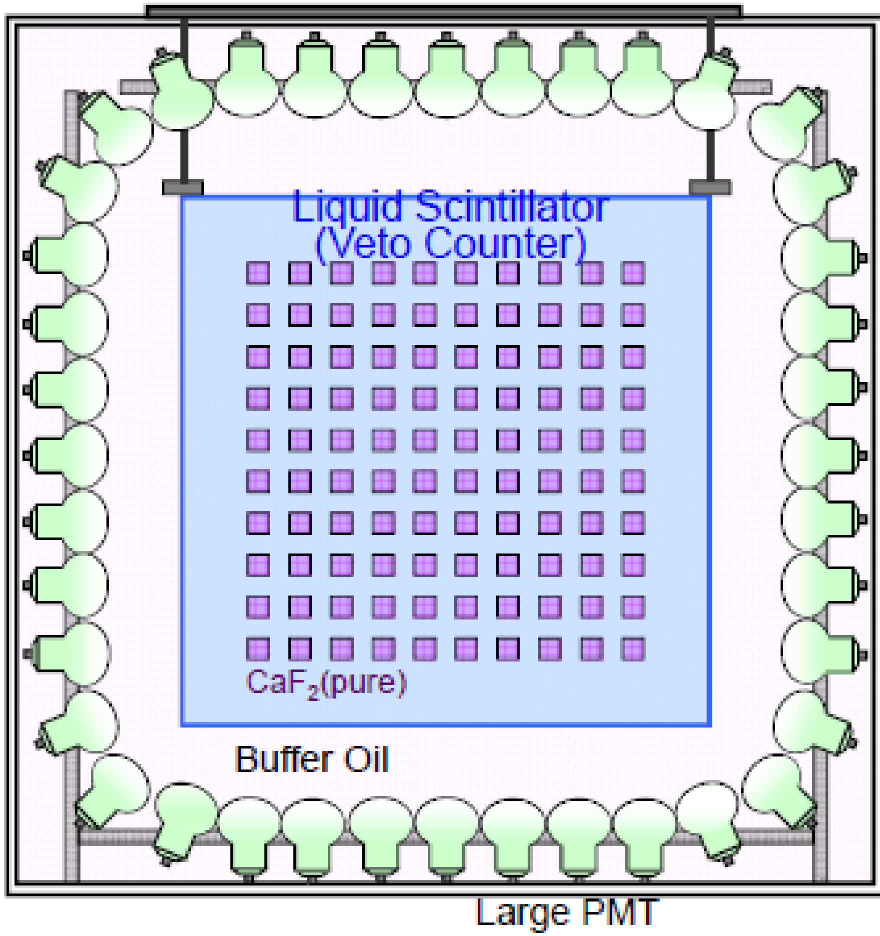
A dream ?

^{48}Ca	CANDLES SuperNEMO AMoRE
^{150}Nd	SNO+ SuperNEMO MTD Borexino

Design Concepts of CANDLES

CANDLES

CAlcium fluoride for studies of Neutrino and Dark matrters by Low Energy Spectrometer



- CaF_2(pure) scintillator
  - Long attenuation length (>10m@350nm)
  - Double beta decay source
  - ^{48}Ca (Q_{bb}=4.27MeV)
- Liquid scintillator
  - 4 π Active Shield
- Large photomultiplier tube
  - Signals from both scintillators are detected simultaneously

⇩

- Active Shielding Technique

Different time constants

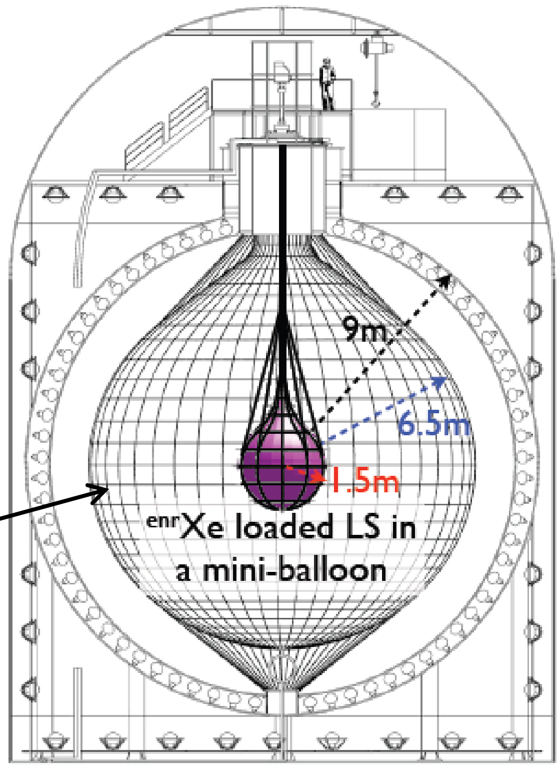
CaF_2(pure) : ~1μsec

Liquid scintillator : a few 10 nsec

UMEHARA Saori, 16th Nov. 2011, DBD11

KamLAND-Zen

Zero Neutrino
double beta decay search



idea to load Xe into LS is from Raju PRL72,1411(1994)

~320kg 90% enriched ^{136}Xe installed so far
total 600+ kg in the mine
production reaches 700kg in this year

Pseudo-cumene 20%
+
Dodecane 80%
+
PPO 0.15wt%

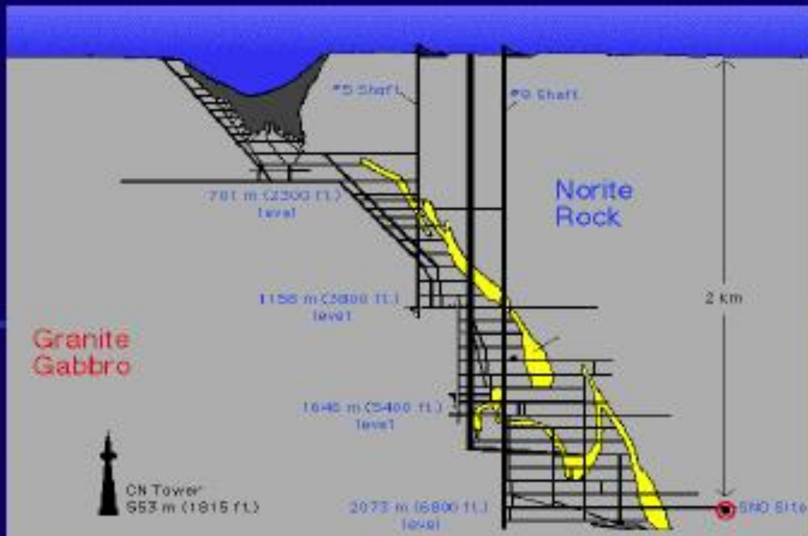
K.Inoue@ν2012

Good features of us

- running detector
  → relatively low cost and quick start
- huge and clean (1200m^3, U: 3.5×10^{-18} g/g, Th: 5.2×10^{-17})
  → negligible external gamma
  (Xe and mini-balloon need to be clean)
- Xe-LS can be purified, mini-balloon replaceable if necessary, with relatively low cost
  → highly scalable (up to several tons of Xe)
- No escape or invisible energy from β, γ
  → BG identification relatively easy
- anti-neutrino observation continues
  → geo-neutrino w/o japanese reactors

Disadvantages toward an ultimate sensitivity

× relatively poor energy resolution
  tolerable thanks to slow 2ν2β and low BG
× no β/γ discrimination so far
× delicate balloon film
× limited LS composition (for density matching)



~~1000 tonnes D_2O~~ → **780 tonnes liquid Scintillator (LAB)**

12 m diameter Acrylic Vessel

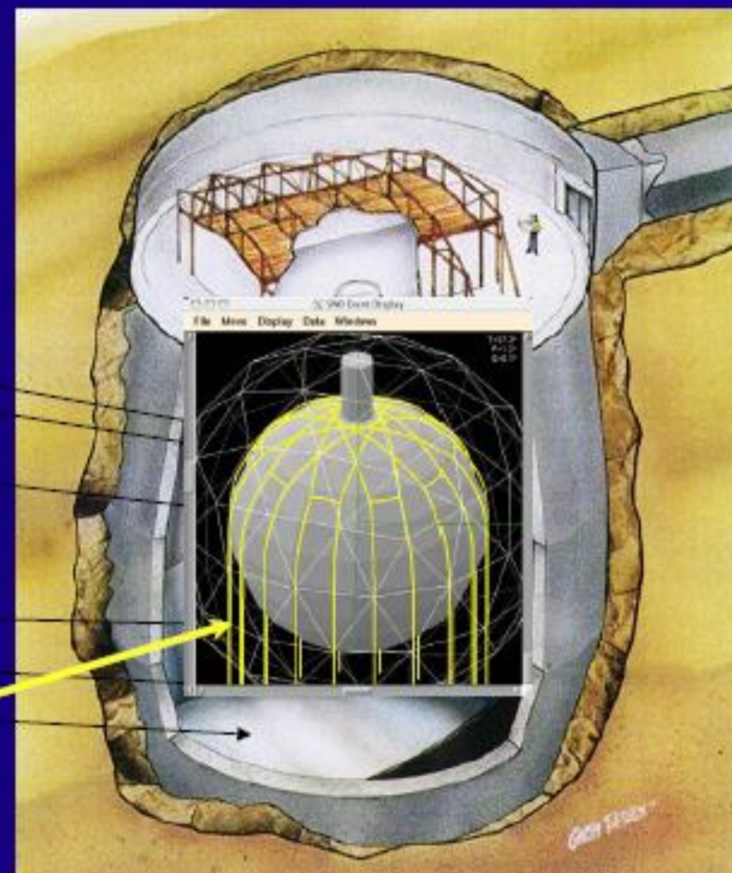
18 m diameter support structure; 9500 PMTs

(~60% photocathode coverage)

1700 tonnes inner shielding H_2O
5300 tonnes outer shielding H_2O
Urylon liner radon seal

hold down rope net

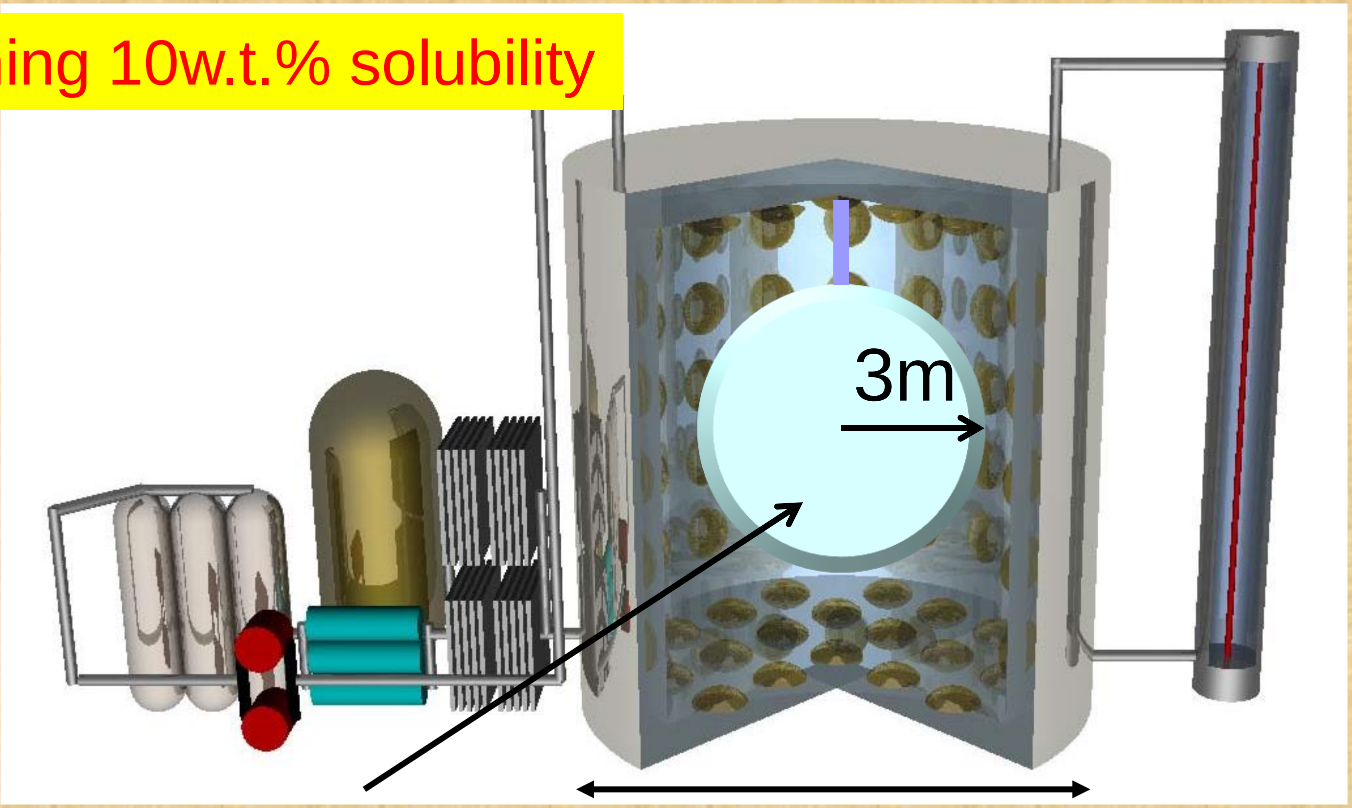
depth: 2092 m (~6010 m.w.e.) ~70 muons/day



Detector design for ZICOS experiment

- **Zirconium Complex in Organic liquid Scintillator (ZICOS) for double beta decay experiment**

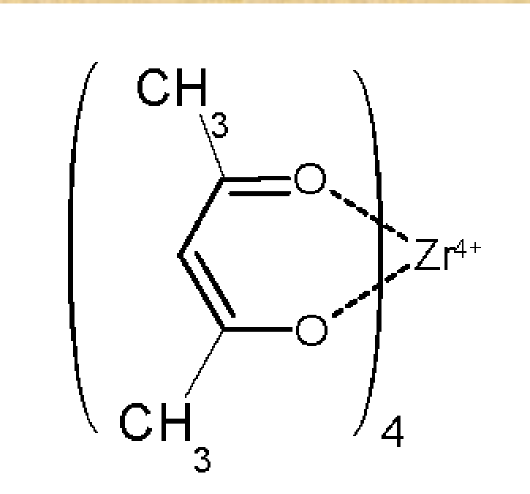
Assuming 10w.t.% solubility



Zirconium loaded 100ton LS

Zirconium β-diketon complex

- Zirconium(IV) acetylacetonate ($Zr(acac)_4$)

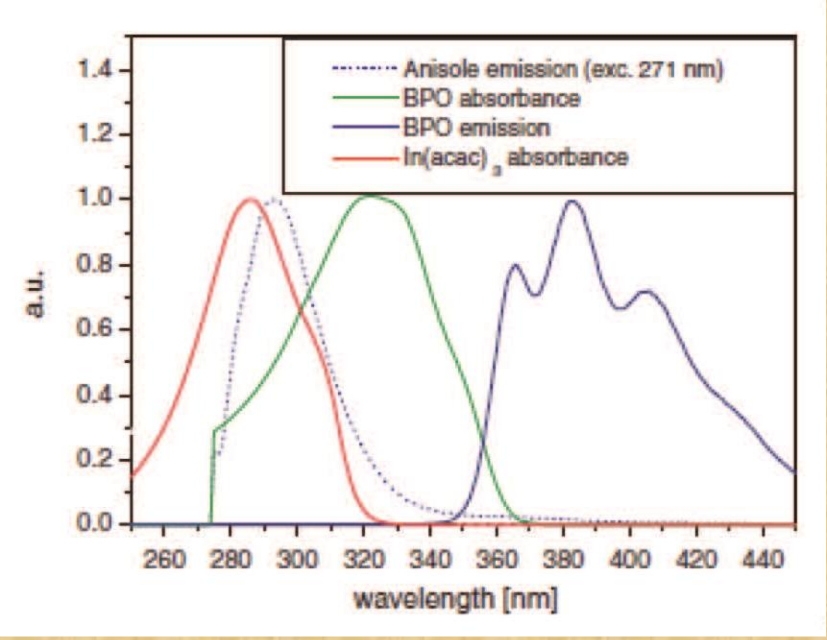


Molecular weight : 487.66

- Advantage
  - good solubility (over 10w.t.%) in Anisole (PhOMe)
  - Stable and cheep
  - Commercial product
- Disadvantage
  - Low scintillation light yield

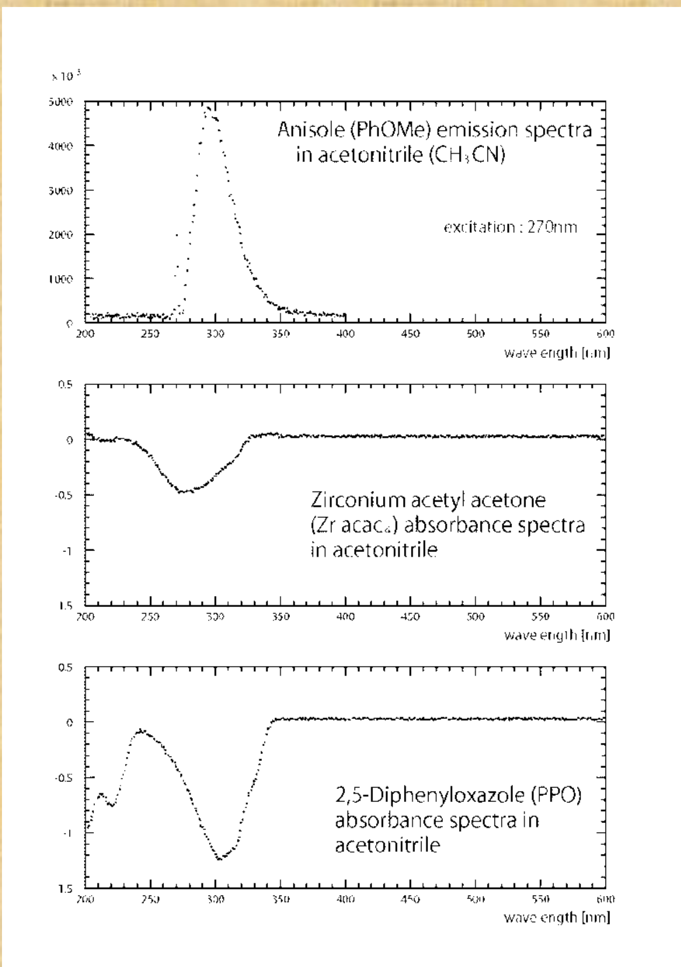
What’s problem

- Absorption spectra of In(acac)$_3$ (indium acetyl acetone) was overlapped with the emission spectra from Anisole（Chem. Phys. Lett., 435(2007), 252)



Same overlap between the emission and the absorption could be occurred even if different metal (Zr) was used.

Observed absorption spectra of $Zr(acac)_4$



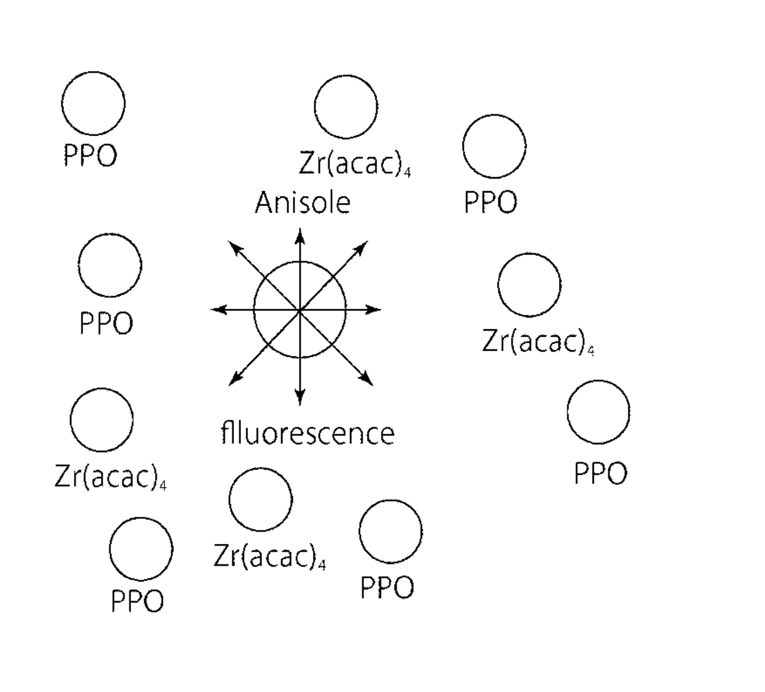
- Emission peak of anisole was observed around 295nm.
- Absorption peak of $Zr(acac)_4$ was observed around 270nm.

Scintillation light from anisole (PhOMe) might be absorbed by $Zr(acac)_4$

Simple expectation for quenching

- Assuming to same cross section for light

$$\text{Light yield} = L_0 \times \frac{\sigma_1 N_{ppo}}{\sigma_1 N_{ppo} + \sigma_2 N_{Zr}}$$

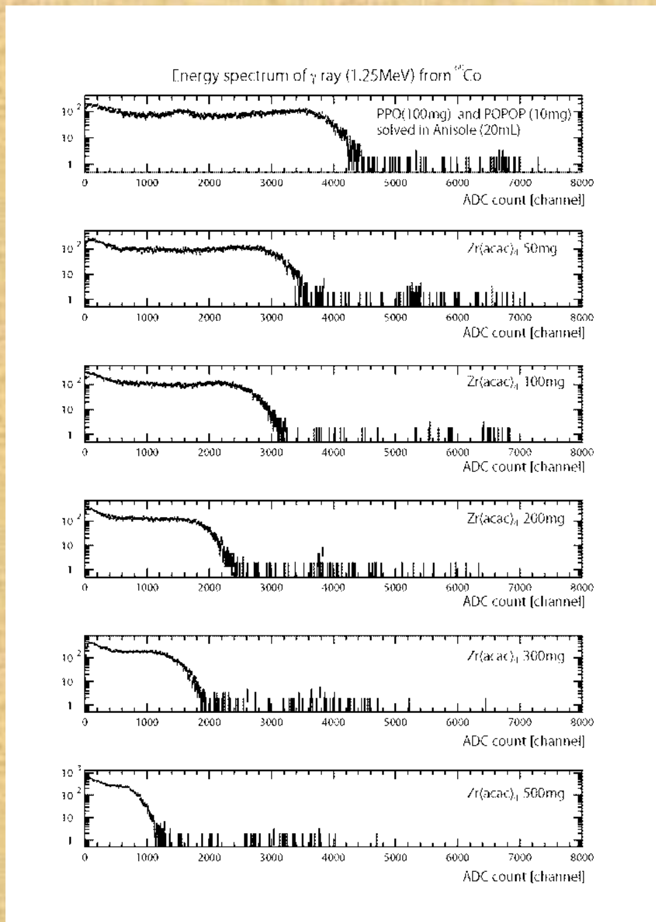


L_0 : Light yield of anisole + PPO + POPOP

N_{ppo} , N_{Zr} : Number of molecular for PPO and $Zr(acac)_4$

σ_1,σ_2 : cross section of absorbance for PPO and $Zr(acac)_4$

Scintillation Light yield (^{60}Co) with respect to concentration of Zr(acac)$_4$

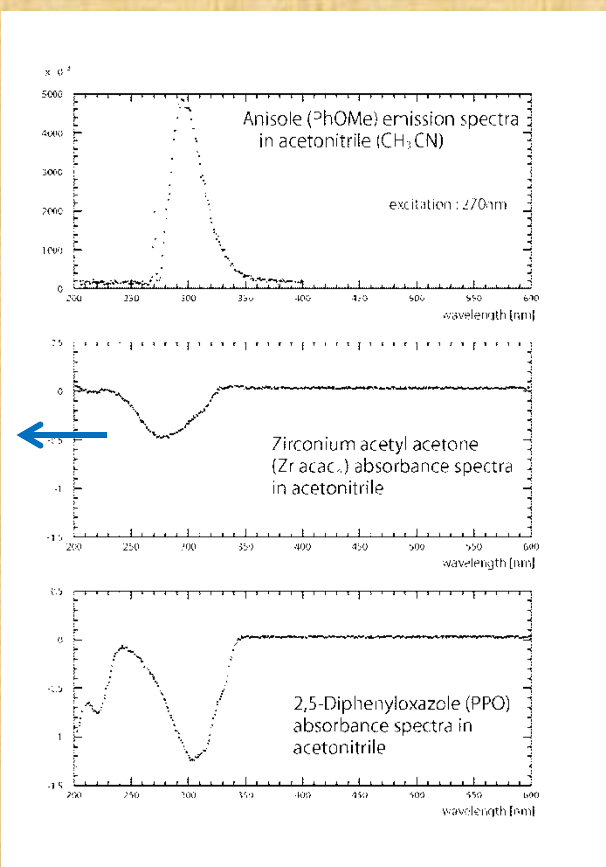


concentration of Zr(acac)$_4$	Observed channel	Expected channel
0 mg	3850	3850
50mg (1.03×10^{-4})	3175	3138
100mg (2.05×10^{-4})	2800	2651
200mg (4.10×10^{-4})	2000	2018
300mg (6.15×10^{-4})	1600	1613
500mg (1.03×10^{-3})	900	1178

PPO 100mg : 4.52×10^{-4} mol

Improvement of scintillation light yield

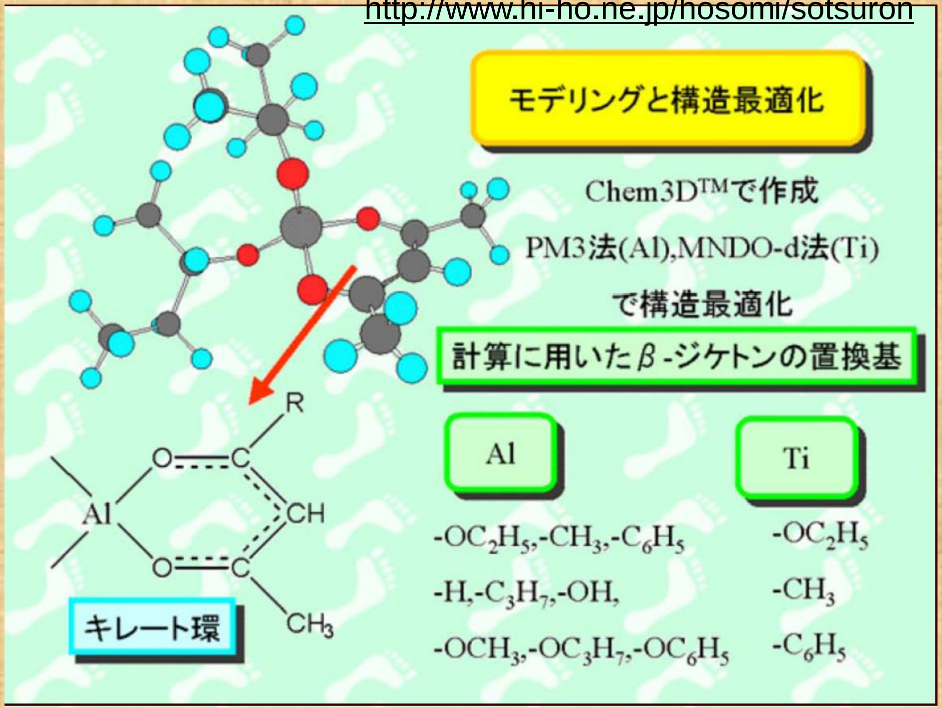
- Move absorption peak to shorter wavelength



- How to do it?

substituent groups

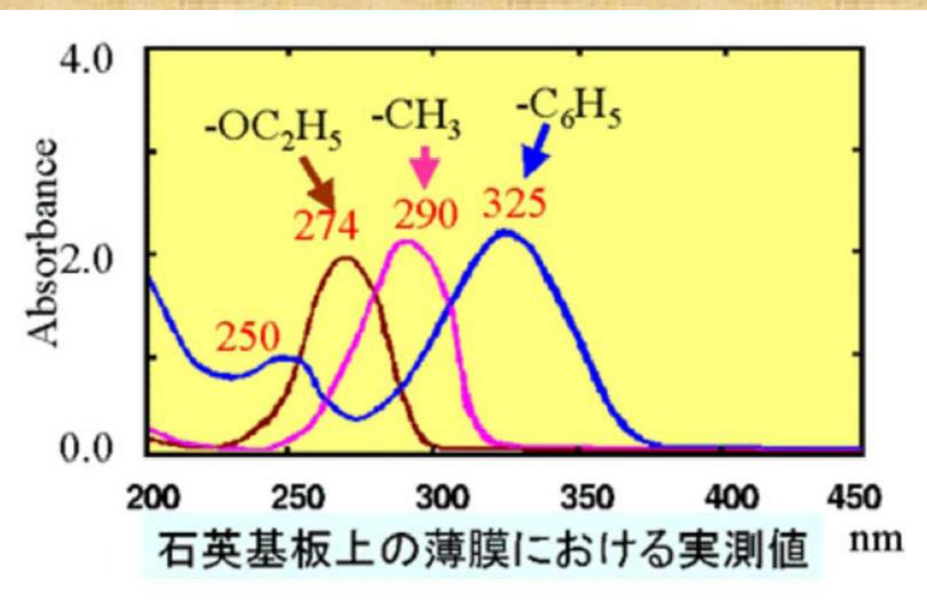
http://www.hi-ho.ne.jp/hosomi/sotsuron

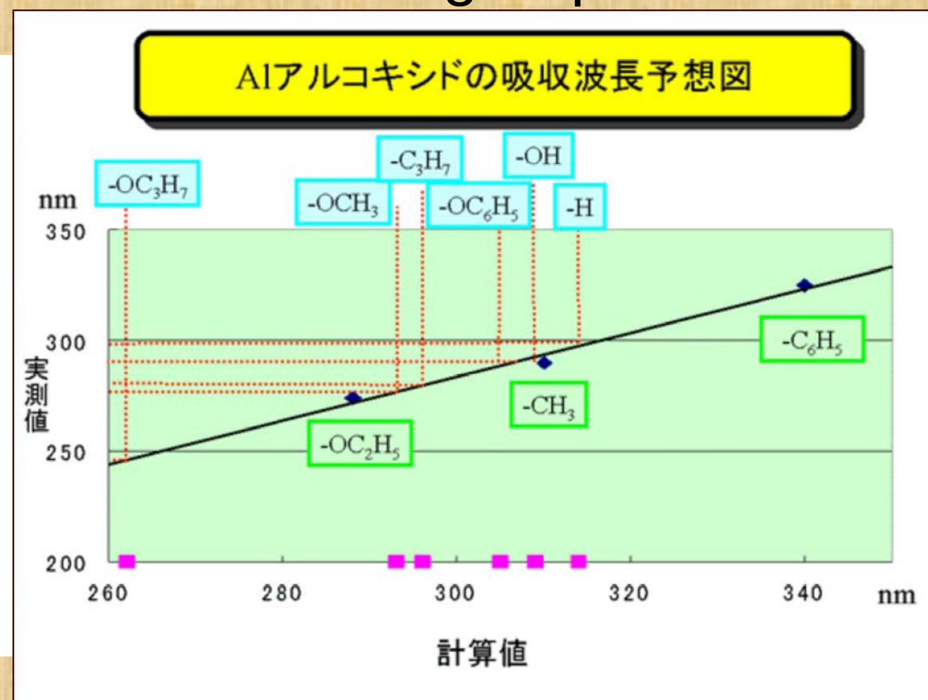


Courtesy of Prof. Yoshiyuki Kowada (Hyogo University of Education)

Absorbance peak for several substituent groups

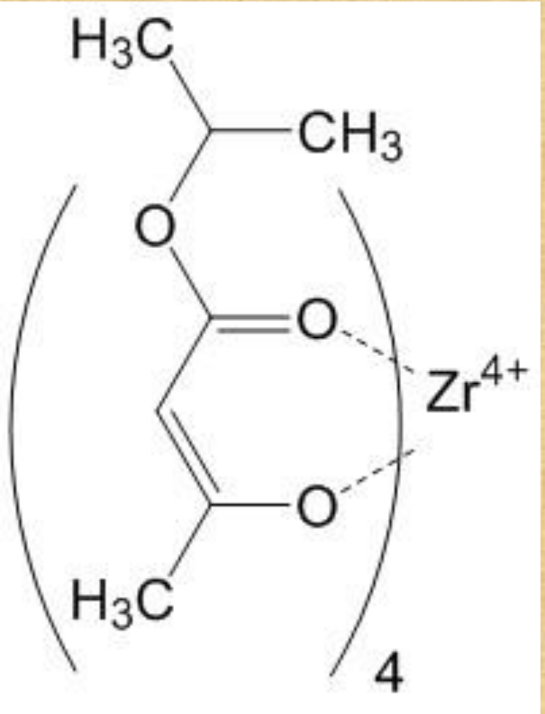
- Measured absorbance peaks for several substituent groups
- Expected absorbance peak for several substituent groups



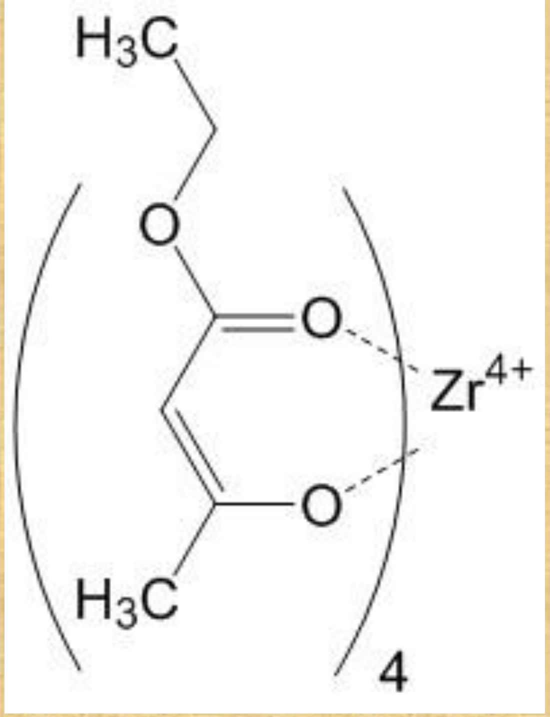


Zr β-diketon complex introducing substituent groups (β-keto ester complex)

Zr(CH3COCHCOOCH (CH3))$_4$ = Zr(iprac)$_4$
mw : 711.92

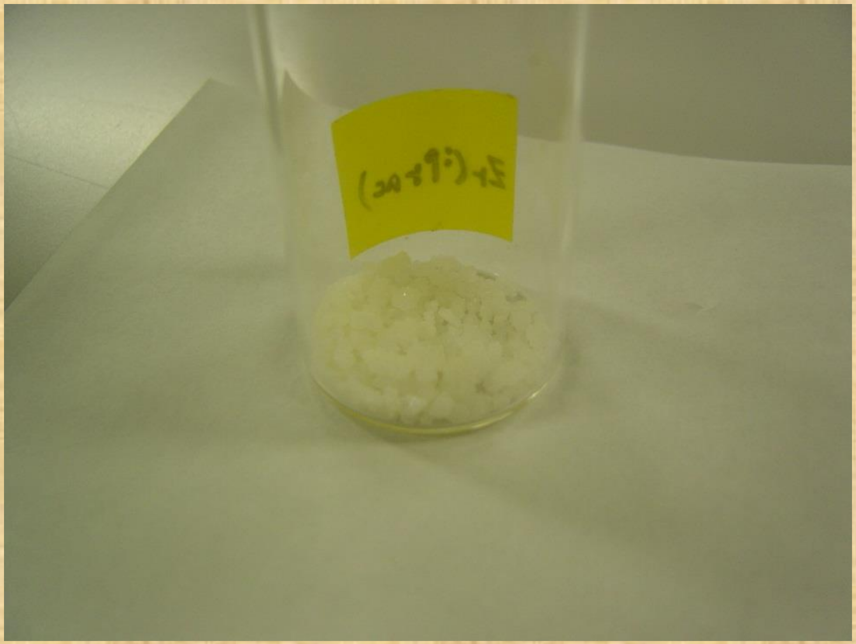


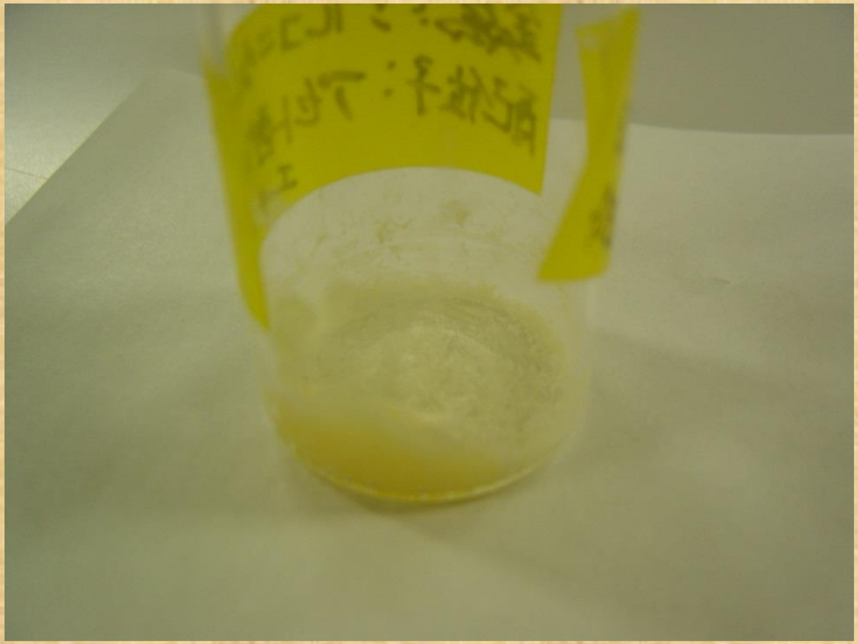
Zr(CH3CCOCHCOO CH3)$_4$ = Zr(etac)$_4$
mw : 665.81



Zr β-keto ester complex

$Zr(iprac)_4$+$(iprac)_{1.5}$	$Zr(etac)_4$
state: powder	state : dry solid



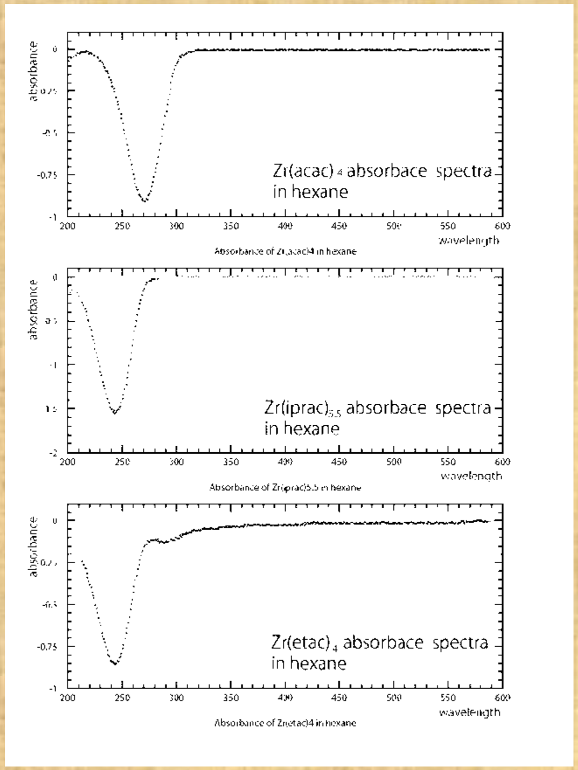


Synthesized by Prof. Takahiro Gunji (Tokyo University of Science)

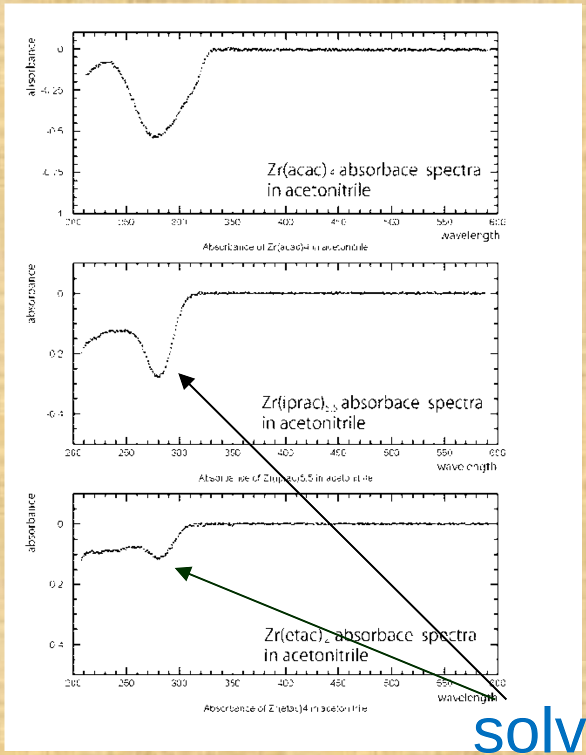
Solubility > 10 w.t.% for anisole

Absorbance spectra (Solvent effect)

Solution : Hexane



Solution : acetonitrile

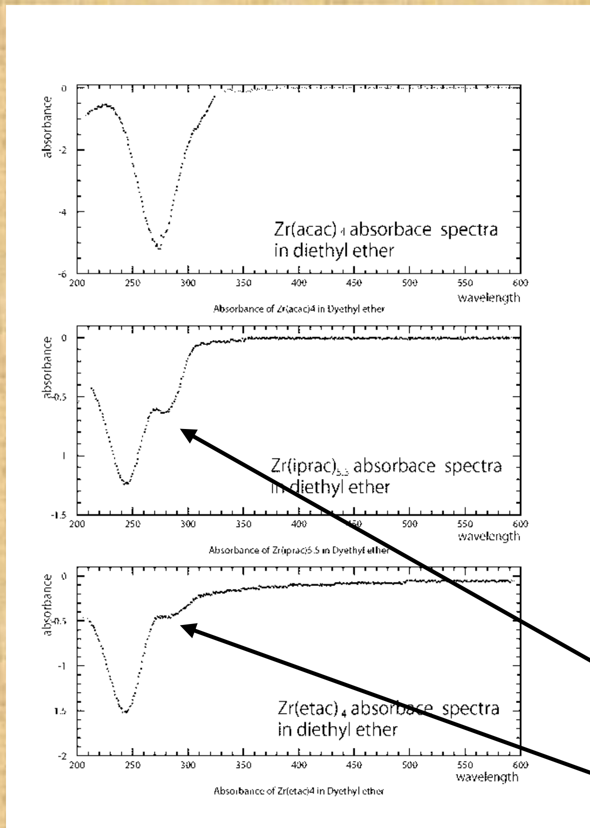


solvent effect

Absorption peak moves to shorter wavelength, however it depends on the polarity of solvent.

Absorbance in liquid scintillator

Solution : Diethyl Ether

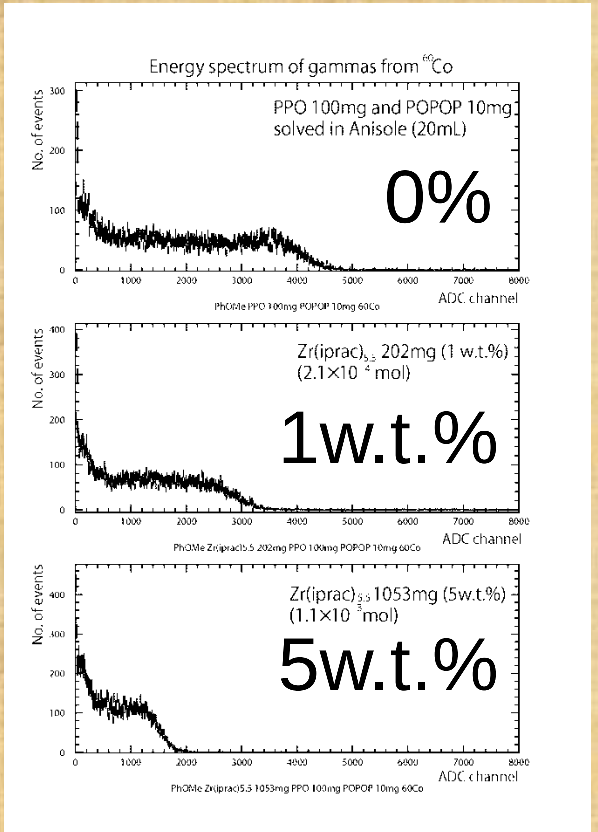


- Solvent effect could depend on the polarity (dielectric const.)
  - Acetonitrile : 37.5
  - Hexane : 1.89
  - Anisole : 4.3
- Need solution which has same polarity as anisole
  - Diethyl ether : 4.33

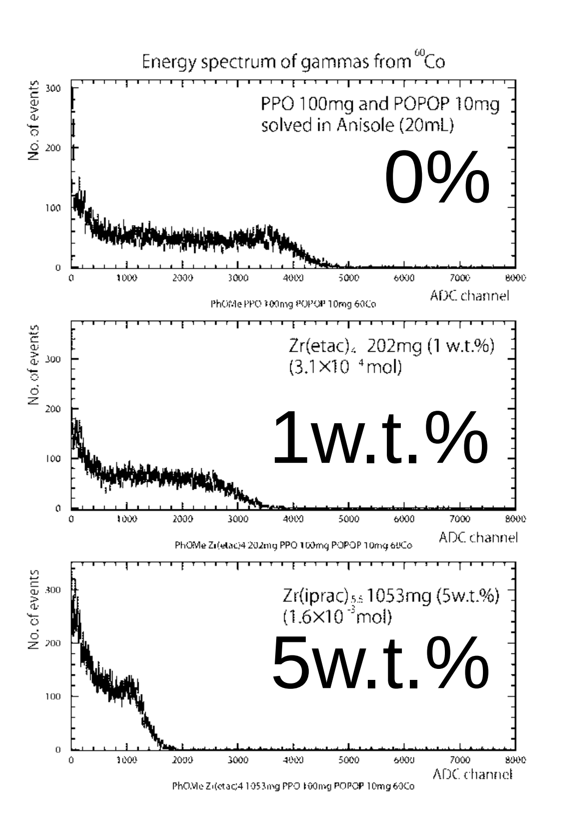
Still solvent effect remains around 270nm

Light yield of Zr β-keto ester scintillator

$Zr(iprac)_{5.5}$ in anisole



$Zr(etac)_4$ in anisole



Same quenching as $Zr(acac)_4$ was observed

Requirement of scintillator solvent

- Low polarity (low dielectric constant)
  - No absorption ~270nm
- Aromatic compounds
  - luminescence >270nm
- Keep high solubility > ~10%
- Keep luminescence from solvent

Possible solvent: Toluene / Xylene

Solvent	mp	bp	D_4^{20}	n_D^{20}	ε	R_D	μ
Acetic acid	17	118	1.049	1.3716	6.15	12.9	1.68
Acetone	-95	56	0.788	1.3587	20.7	16.2	2.85
Acetonitrile	-44	82	0.782	1.3441	37.5	11.1	3.45
Anisole	-3	154	0.994	1.517	4.33	33	1.38
Benzene	5	80	0.879	1.5011	2.27	26.2	0
Bromobenzene	-31	156	1.495	1.558	5.17	33.7	1.55
Carbon disulfide	-112	46	1.274	1.6295	2.6	21.3	0
Carbon tetrachloride	-23	77	1.594	1.4601	2.24	25.8	0
Chlorobenzene	-46	132	1.106	1.5248	5.62	31.2	1.54
Chloroform	-64	61	1.489	1.4458	4.81	21	1.15
Cyclohexane	6	81	0.778	1.4262	2.02	27.7	0
Dibutyl ether	-98	142	0.769	1.3992	3.1	40.8	1.18
o -Dichlorobenzene	-17	181	1.306	1.5514	9.93	35.9	2.27
1,2-Dichloroethane	-36	84	1.253	1.4448	10.36	21	1.86
Dichloromethane	-95	40	1.326	1.4241	8.93	16	1.55
Diethylamine	-50	56	0.707	1.3864	3.6	24.3	0.92
Diethyl ether	-117	35	0.713	1.3524	4.33	22.1	1.3
1,2-Dimethoxyethane	-68	85	0.863	1.3796	7.2	24.1	1.71
N,N -Dimethylacetamide	-20	166	0.937	1.4384	37.8	24.2	3.72
N,N -Dimethylformamide	-60	152	0.945	1.4305	36.7	19.9	3.86
Dimethyl sulfoxide	19	189	1.096	1.4783	46.7	20.1	3.9
1,4-Dioxane	12	101	1.034	1.4224	2.25	21.6	0.45
Ethanol	-114	78	0.789	1.3614	24.5	12.8	1.69
Ethyl acetate	-84	77	0.901	1.3724	6.02	22.3	1.88
Ethyl benzoate	-35	213	1.05	1.5052	6.02	42.5	2
Formamide	3	211	1.133	1.4475	111	10.6	3.37
Hexamethylphosphoramide	7	235	1.027	1.4588	30	47.7	5.54
Isopropyl alcohol	-90	82	0.786	1.3772	17.9	17.5	1.66
Methanol	-98	65	0.791	1.3284	32.7	8.2	1.7
2-Methyl-2-propanol	26	82	0.786	1.3877	10.9	22.2	1.66
Nitrobenzene	6	211	1.204	1.5562	34.82	32.7	4.02
Nitromethane	-28	101	1.137	1.3817	35.87	12.5	3.54
Pyridine	-42	115	0.983	1.5102	12.4	24.1	2.37
Tetrahydrofuran	-109	66	0.888	1.4072	7.58	19.9	1.75
Toluene	-95	111	0.867	1.4969	2.38	31.1	0.43
Trichloroethylene	-86	87	1.465	1.4767	3.4	25.5	0.81
Triethylamine	-115	90	0.726	1.401	2.42	33.1	0.87
Trifluoroacetic acid	-15	72	1.489	1.285	8.55	13.7	2.26
2,2,2-Trifluoroethanol	-44	77	1.384	1.291	8.55	12.4	2.52
Water	0	100	0.998	1.333	80.1	3.7	1.82
p -Xylene	-25	144	0.88	1.5054	2.57	35.8	0.62

Future Solar Neutrino Experiments (Beyond those already in operation)

pep/CNO	Medium	Status
SNO+	780 kg LAB Liq scintillator	Construction, start 2013
Kamland-2	780 lb Liq Scintillator	Following KamLAND-Zen

For pp, ^{7}Be neutrinos, measuring CC plus ES could extract electron and total neutrino fluxes

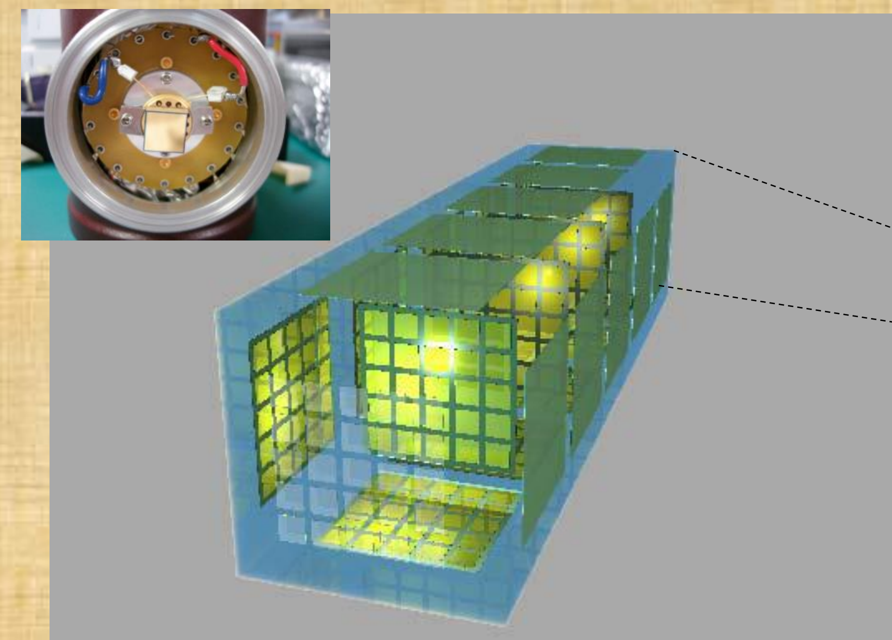
pp via ES		
XMASS	20 tons Liq Xe	835 kg since 2010 for $\beta\beta$
CLEAN	50 tons Liq Ne	MiniClean (500 kg) start 2013
P, ^{7}Be via CC		
LENS	10 tons ^{115}In	μLENS under development
MOON	3 tons ^{100}Mo	R&D in progress
IPNOS	^{115}In	R&D in progress
MEGAPROJECTS	Threshold defines: ^{8}B + ?	
HyperK, MEMPHYS	Megaton Water Cerenkov	
LBNE, GLACIER	50 to 100 kTon Liquid Ar	
LENA	50 kTon Liq Scintillator	

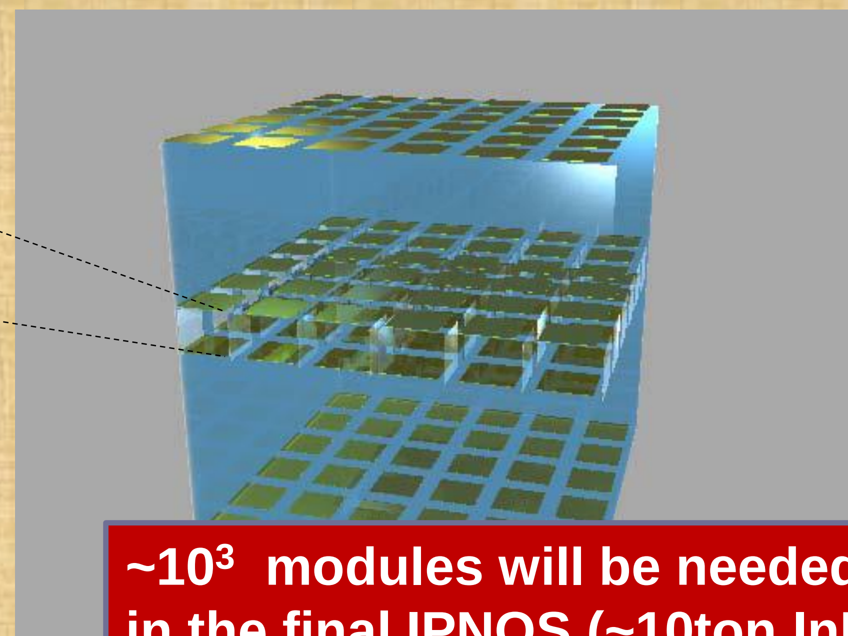
A.McDonald @ ν2012

IPNOS phase-I experiment for low energy solar ν experiment

InP multi-pixel detector inside of Liquid Xenon (LXe) with PMTs

30cm cubic chamber (like XMASS 100kg prototype) includes ~10kg InP detector

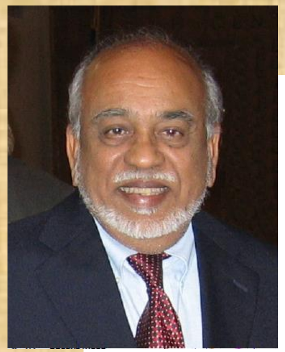




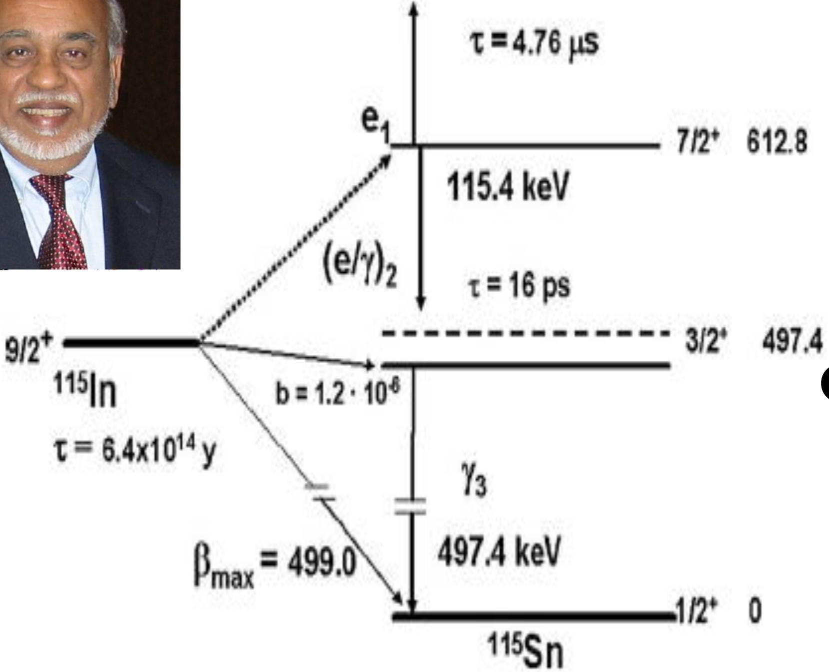
~10^3 modules will be needed in the final IPNOS (~10ton InP)

Need larger area InP detector

Capture of low energy solar neutrinos by ^{115}In



R.S.Raghavan Phs.Rev.Lett37(1976)259



Nuclear Physics A 748 (2005) 333-347

$^{115}In + \nu_e \rightarrow {}^{115}Sn^* + e^-$

$^{115}Sn^*(4.76\mu s) \rightarrow {}^{115}Sn +$

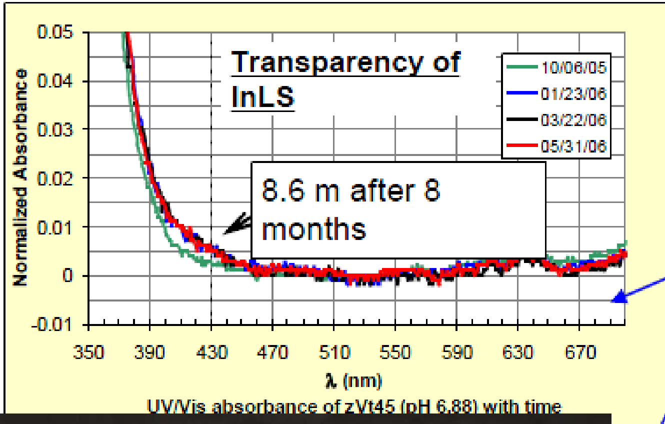
$\gamma_1(115keV) + \gamma_2(497keV)$

● Advantage
- large cross section (~640SNU)
- direct counting for solar neutrinos
- sensitive to low energy region ($E_\nu \geqq$ 125keV)
- energy measurement ($E_e = E_\nu -$ 125keV)
- triple fold coincidence to extract neutrino signal from huge BG ($e_1 + \gamma_2 + \gamma_3$)

● Disadvantage
- natural β-decay of ^{115}In ($\tau_{1/2} = 4.4 \times 10^{14}$ yr , Ee≧498keV)
- possible BG due to correlated coincidence by radiative Bremsstrahlung

Goal

1. Good energy resolution : 10%(FWHM)
2. Fine segmentation (10^4-10^5)
3. High efficiency γ detection



Technology and Bgd Control

< Towards **Hi Precision** fluxes >

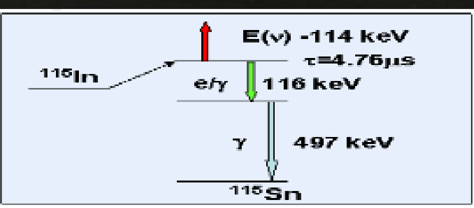
LENS

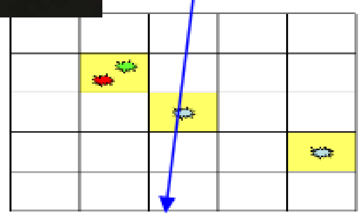
R. Raghavan@Neutrino2010

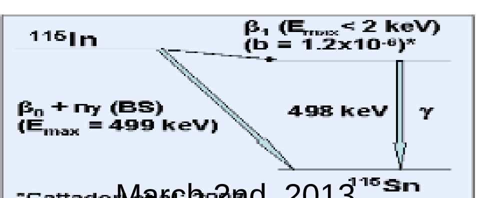
- **Hi Quality InLS**
- **Granular Detector Design**
- **Background Analysis Insights**

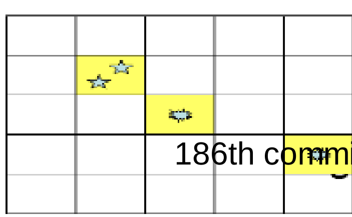
•→ In decay bgd suppressed →S/N ~3 for first time

	Status
Design of Detector	Cubic Lattice Chamber
InLS: In content Light attenuation L(1/e) Signal Eff Pe/MeV	>8% >8m 900
Indium Mass(1900 pp/5y)	10 ton
Total Mass	125 ton
PMT's	13,300
Neutrino detection eff.	64%



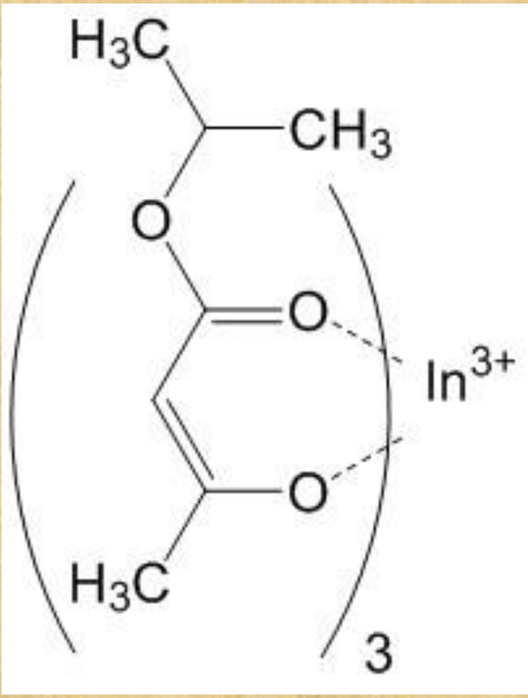






Development for new LS for IPNOS phase-II (solar neutrino experiment)

- Liquid Scintillator containing Inium β-keto ester complex

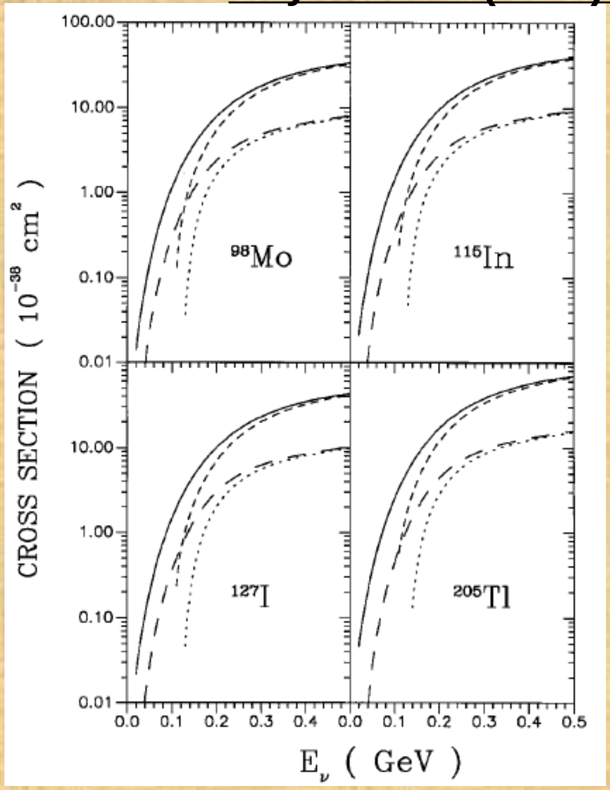


- Advantages
  - Cheep and stable
  - 10ton In in 100ton LS
  - Possible same design as ZICOS
- Low energy solar ν
  - pp/^{7}Be and CNO ν
  - Modulation using ^{7}Be ν
- Supernova ν burst
  - ν_e burst form Neutralization

Cross section for 100MeV- 1GeV ν

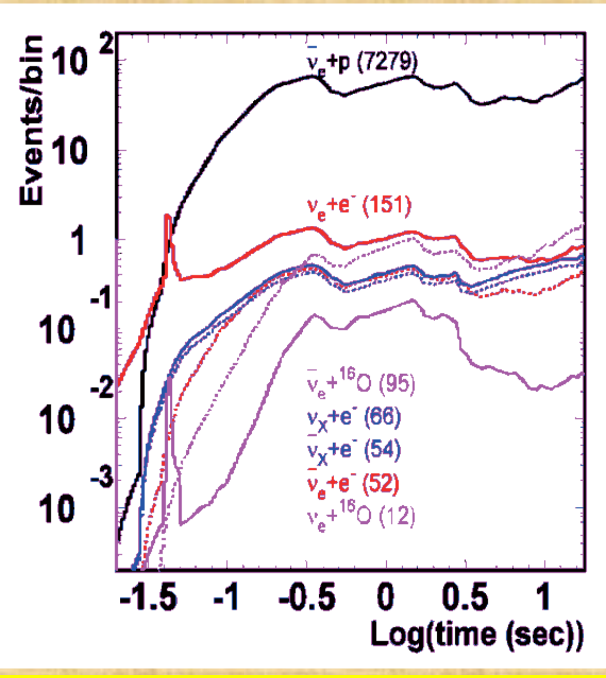
- ν-nucleus cross section

Rhy.Rev.C53(1996)1409



^{115}In has ~10^{-39} cm^2 for 20-100Me compared with $\sigma_{\nu ee}$~10^{-43} cm^2

- Sensitive to neutralization burst

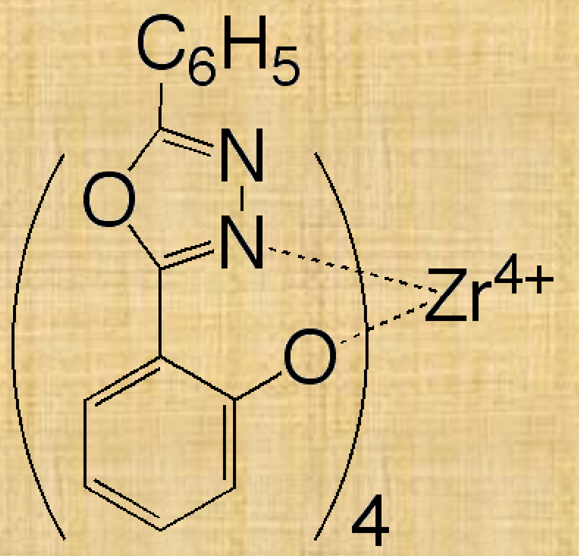


100ton of indium detects 20-100 ev

- Nuclear synthesis (r-process)
- ν mass hierarchy

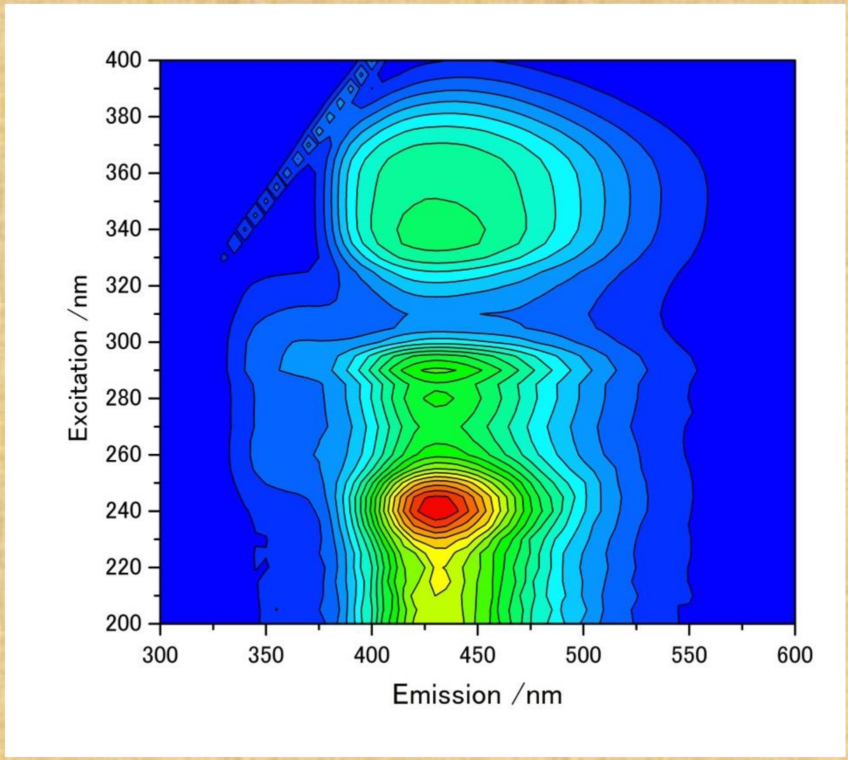
Zirconium complex with luminescence

- Zr-ODZ complex



m.w. = 1040.18

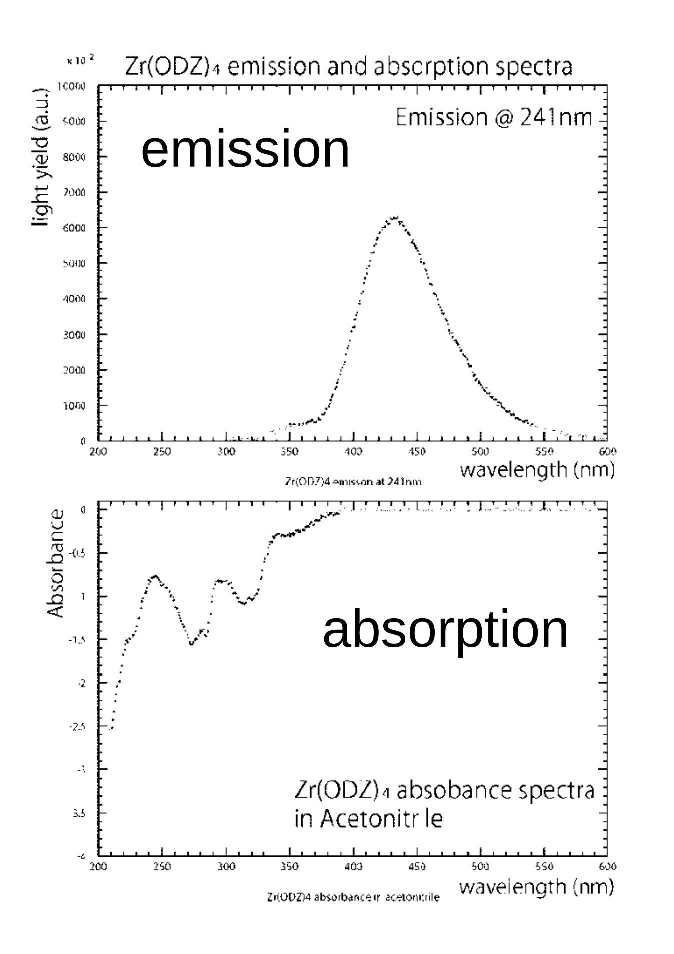
- Photo luminescence



➤**Solvent : Acetonitrile**

➤**Concentration : 3.0×10^{-5} mol/L**

Emission and absorption spectrum of $Zr(ODZ)_4$



- Emission wavelength : 430nm

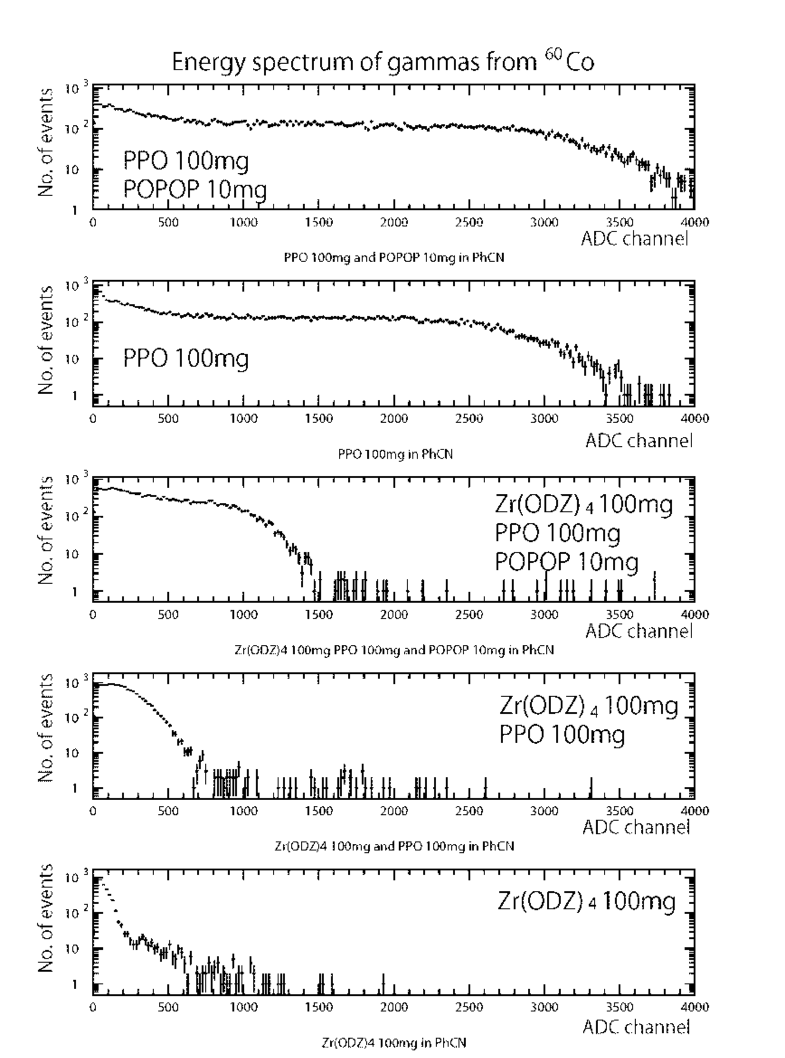
PMT sensitive

- Absorption wavelength: 270nm and 320nm

different from excitation W.L.

- Solvent : PhCN (Benzonitrile)
- Solubility : ~5w.t.%

Response for γ-irradiation



- Most of emission light from PhCN was not used for the emission of $Zr(ODZ)_4$.
- third excitation of ~340nm from PPO was used for the emission of $Zr(ODZ)_4$.
- Estimated Quantum yield was obtained ~30% at first excitation of ~240nm.

Need another solvent which has shorter emission wavelength than PhCN.

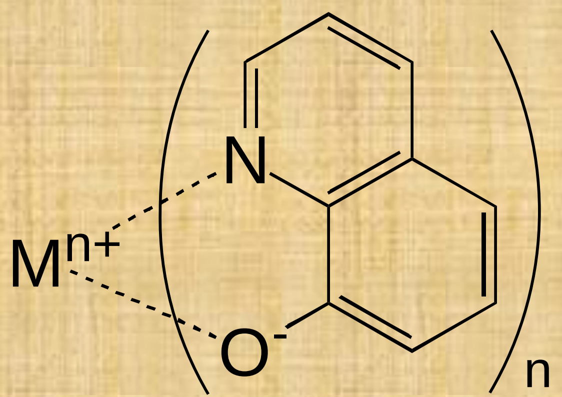
Summary

- Liquid scintillator is ordinal and popular technique to observe neutrinos
- High solubility of Zirconium β-keto ester in Anisole (>~10w.t.%) was achieved.
- Confirmed absorption peak moves to shorter wavelength (275nm → 245nm) by introducing ester substituent groups.
- Observed scintillation light yield decreased in proportion to the concentration of Zr β-keto ester due to solvent effect ⇒ Need low polarity solvent
- Indium β-keto ester complex will be developed for IPNOS phase-II in next fiscal year.

BACKUP

Tetrakis 8-quinolinolate Zr complex loaded scintillator

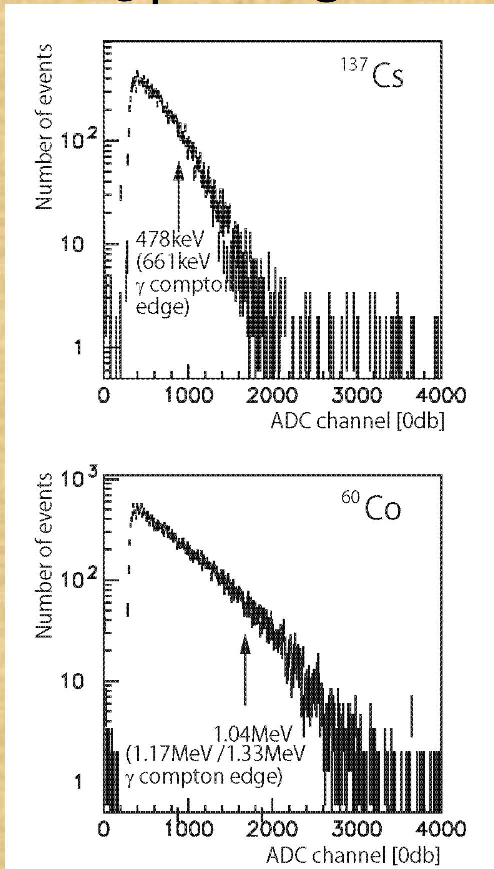
- Tetrakis (8-quinolinolate) Zirconium complex (ZrQ_4)



M = In, n = 3; M = Zr, n = 4

ZrQ_4 m.w. = 689.07

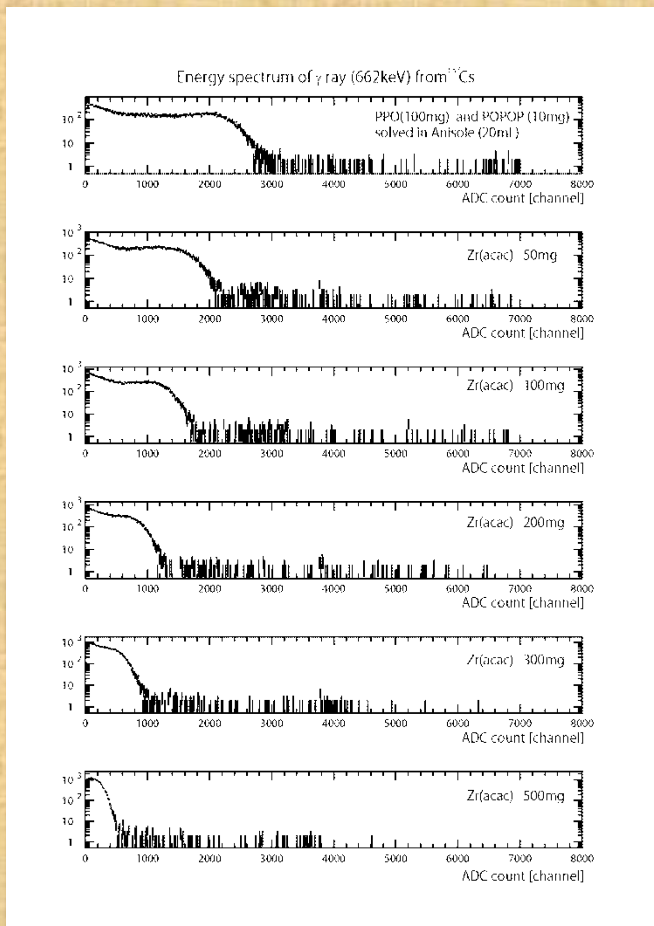
ZrQ_4 50mg in PhCN-POPOP



Quantum Yield=1.1% obtained by optical method

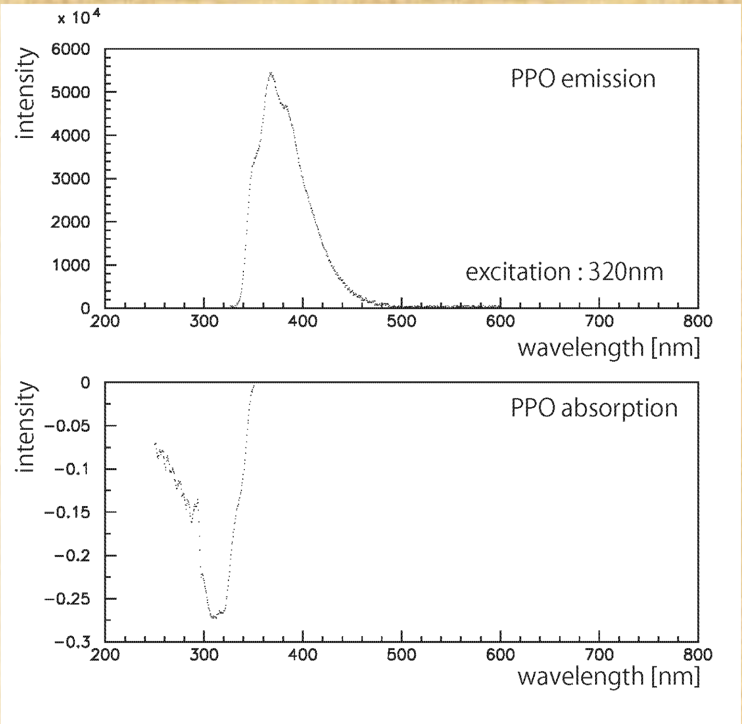
Light Yield to BC505: =7.3%

Scintillation light yield (^{137}Cs) with respect to concentration of $Zr(acac)_4$



concentration of $Zr(acac)_4$	**Observed channel**	**Expected channel**
0 mg	2450	2450
50mg	1800	1997
100mg	1400	1687
200mg	950	1284
300mg	650	1038
500mg	300	750

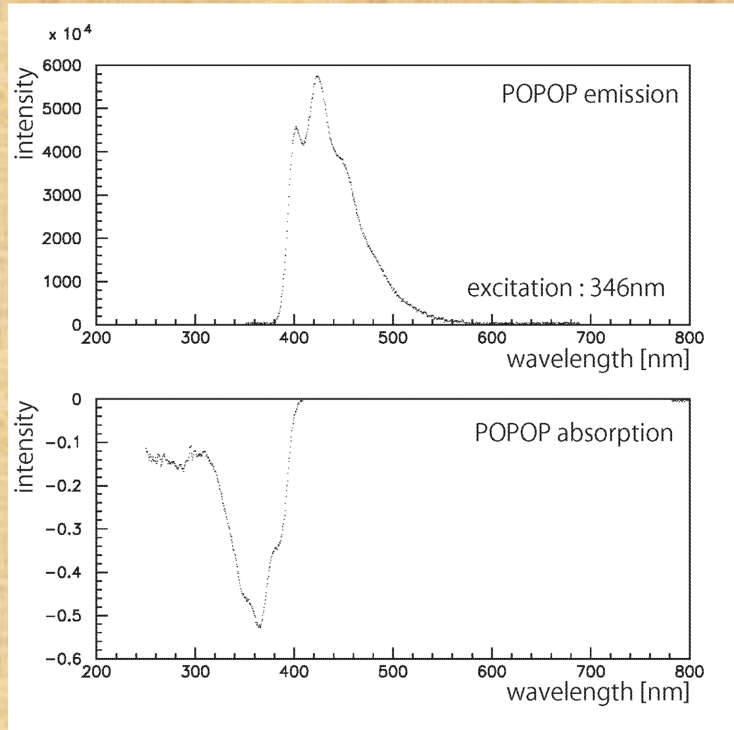
Photo Luminescence and absorption of PPO



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L

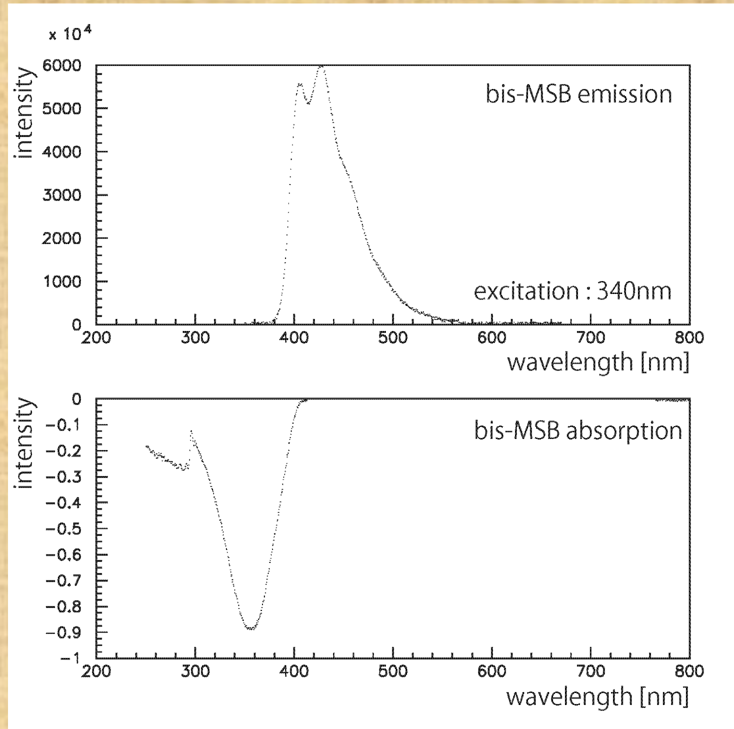
  - 2,5-Diphenyloxazole
  - Molecular mass : 221.26
  - Max. emission wavelength : 368.0nm
  - Max. absorption wavelength : 309.7nm

Photo Luminescence and absorption of POPOP



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L
  - 1,4-Bis(5-phenyloxazol-2-yl)benzene
  - Molecular mass : 364.40
  - Max. emission wavelength : 423.6nm
  - Max. absorption wavelength : 364.1nm

Photo Luminescence and absorption of bis-MSB



- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L

  - 1,4-Bis(2-methylstyryl)benzene
  - Molecular mass : 310.44
  - Max. emission wavelength : 426.6nm
  - Max. absorption wavelength : 355.3nm