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ジルコニウム96を用いたニュートリノを放出しない2重ベータ崩壊事象の探索実験II

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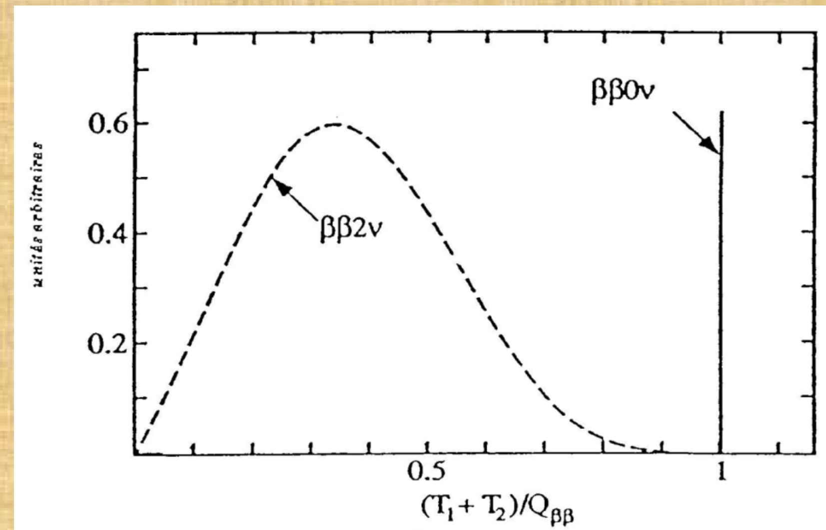
Neutrinoless double beta decay

$\beta\beta$ 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



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

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

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

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

Studied isotopes

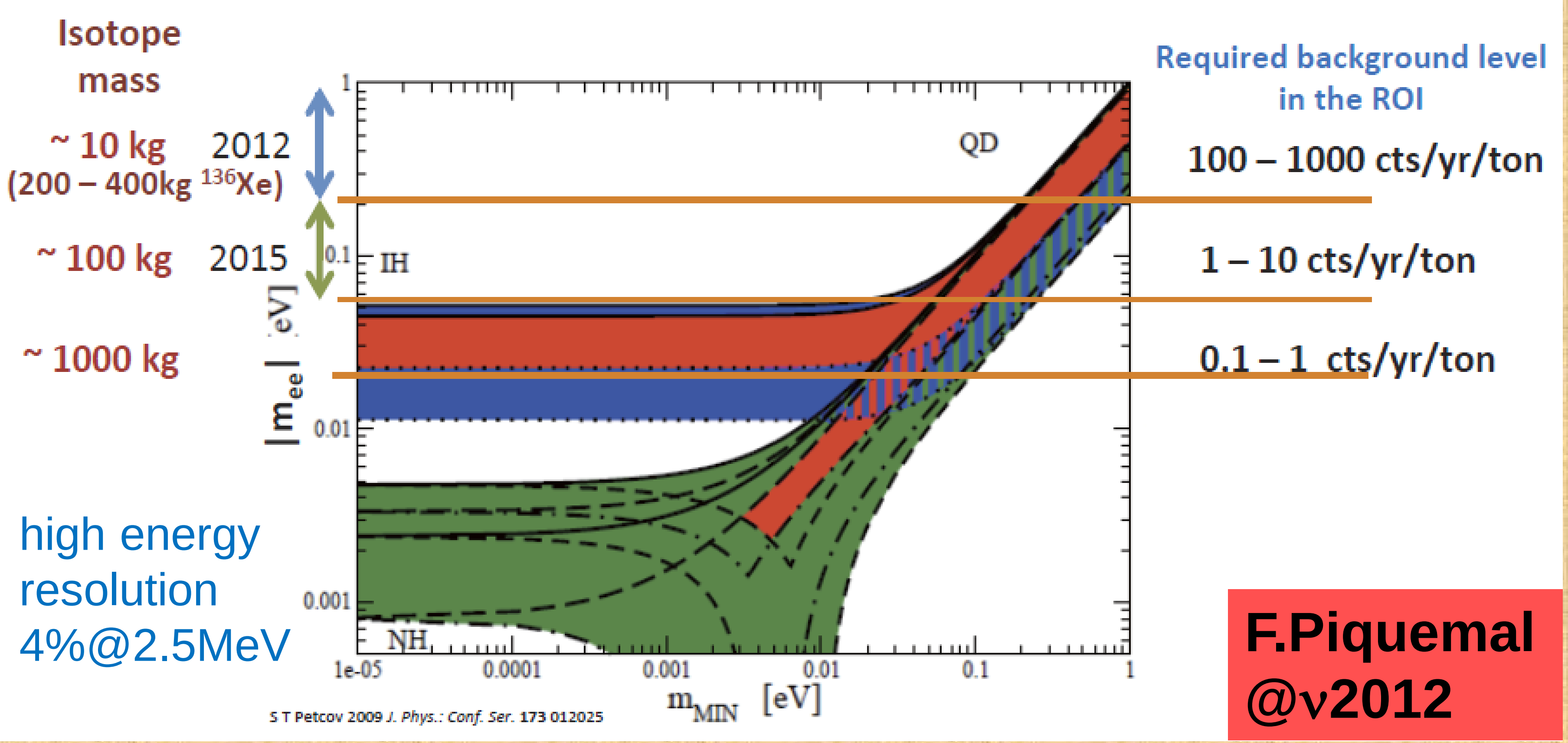
Piquemal@ν2012

- ^{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

For future experiments



high energy resolution 4%@2.5MeV

F.Piquemal @ν2012

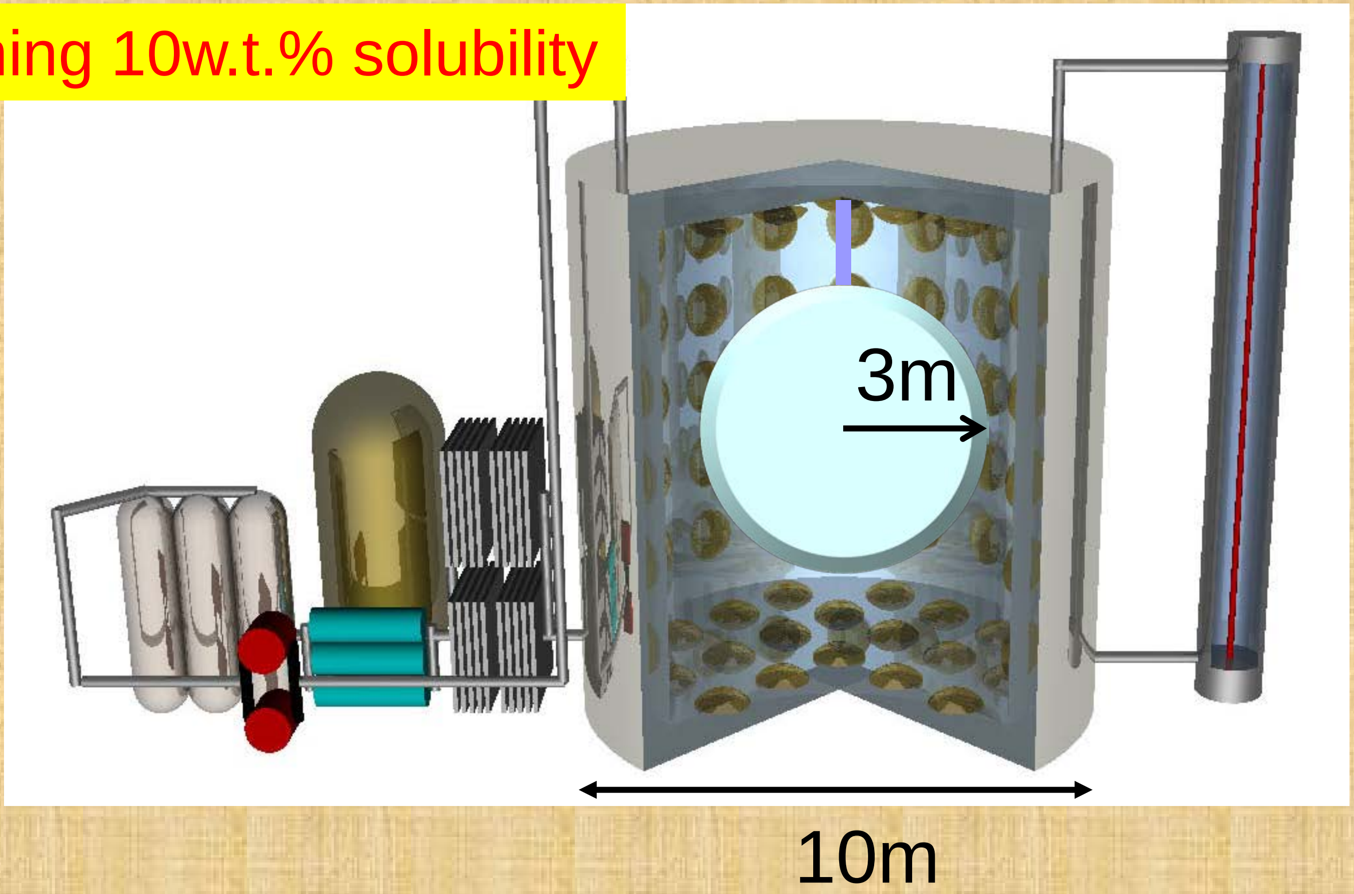
http://kds.kek.jp/getFile.py/access?contribId=37&sessionId=16&resId=2&materialId=slides&confId=9151

~tons of target will be needed for next generation detector

Detector design for Zr in 100ton LS

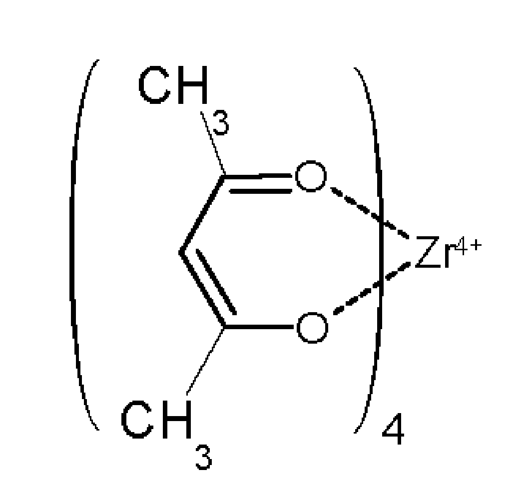
- **Zirconium Complex in Organic liquid Scintillator (ZICOS)**

Assuming 10w.t.% solubility



Zirconium β-diketon complex

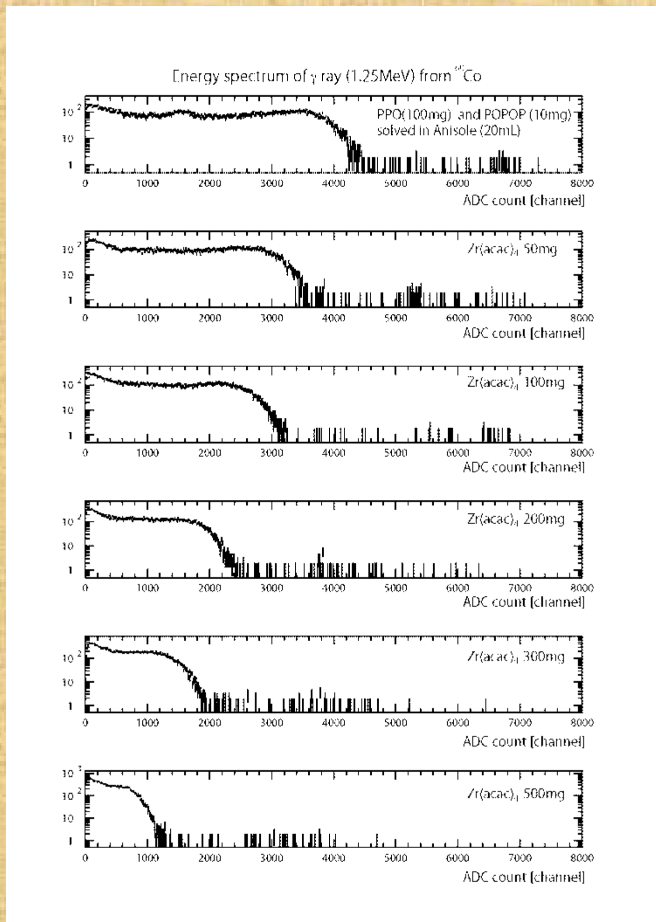
- Zirconium(IV) acetylacetonate ($Zr(acac)_4$)



Molecular weight : 487.66

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

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

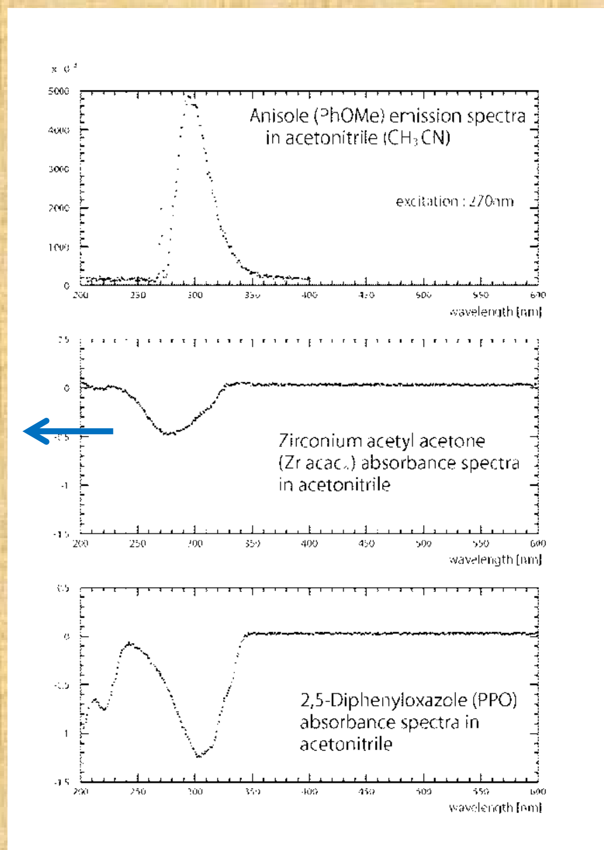


concentration of $Zr(acac)_4$	Observed channel	Expected channel
0 mg	3850	3850
50mg ($1.03X10^{-4}$)	3175	3138
100mg ($2.05X10^{-4}$)	2800	2651
200mg ($4.10X10^{-4}$)	2000	2018
300mg ($6.15X10^{-4}$)	1600	1613
500mg ($1.03X10^{-3}$)	900	1178

PPO 100mg : $4.52X10^{-4}$ mol

Improve scintillation light yield

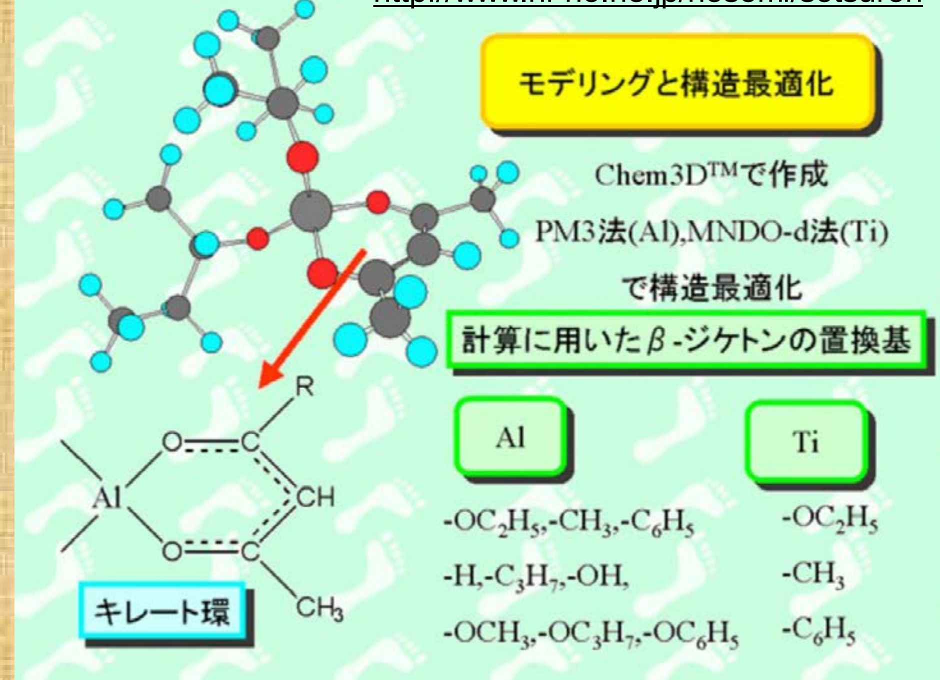
- Move absorption peak to shorter wavelength



- How to do it?

  substituent groups

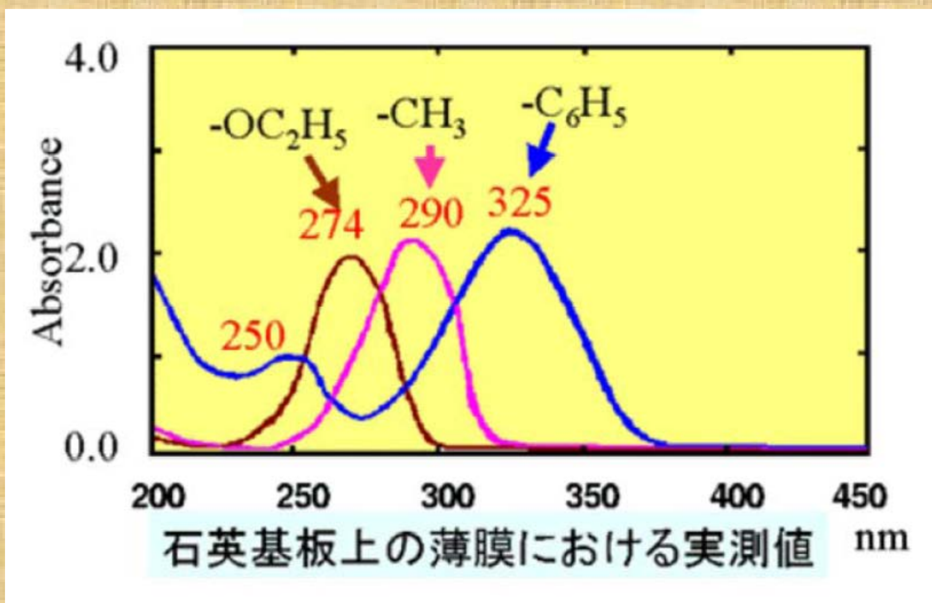
http://www.hi-ho.ne.jp/hosomi/sotsuron



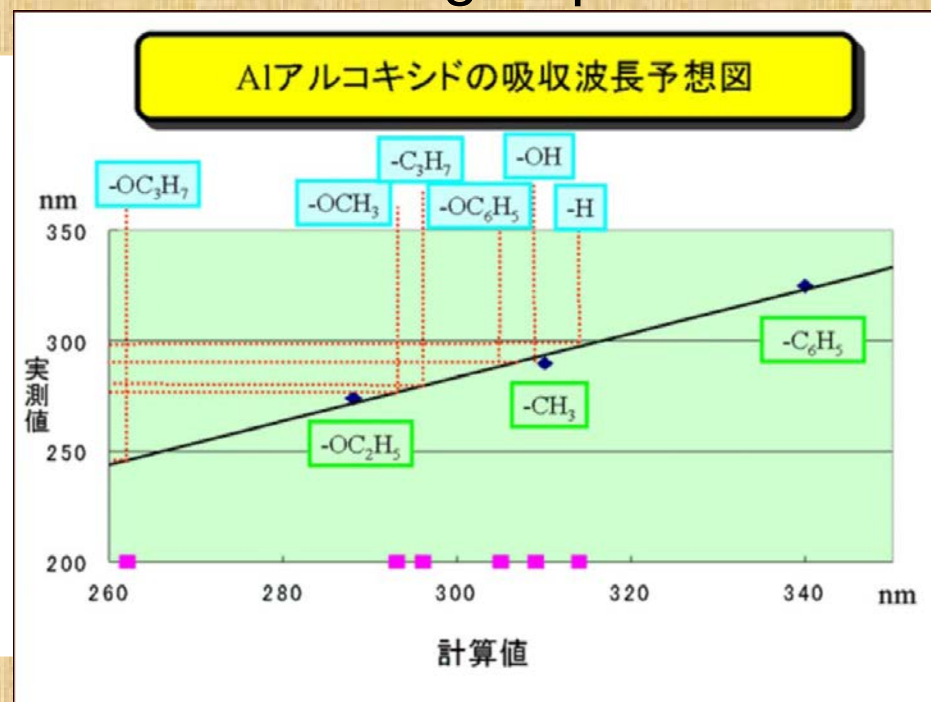
Courtesy of Prof. Yoshiyuki Kowada (Hyogo University of Education)

Absorbance peak for several substituent groups

- Measured absorbance peaks for several substituent groups

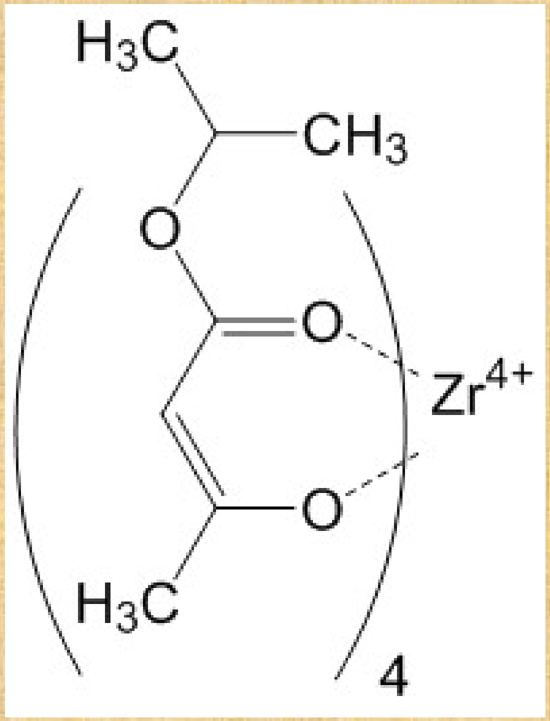


- Expected absorbance peak for several substituent groups

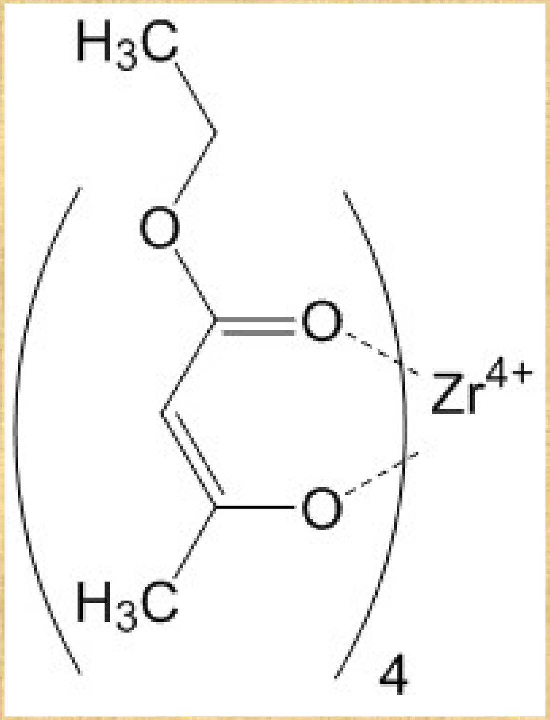


Zr β-diketon complex introducing substituent groups (β-keto ester complex)

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



Zr(CH3CCOCHCOOCH3)$_4$ = Zr(etac)$_4$
mw : 665.81



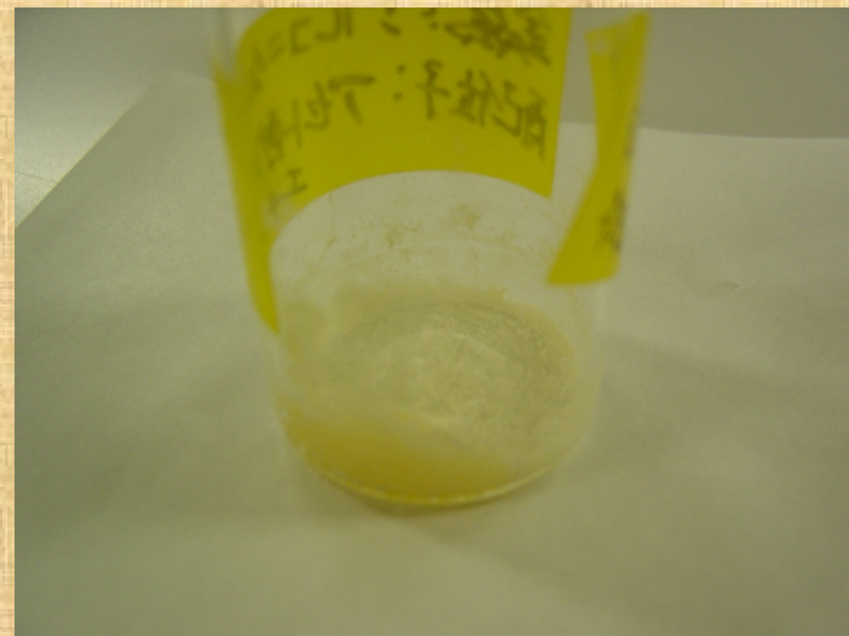
Zr β-keto ester complex

Zr(iprac)$_4$+(iprac)$_{1.5}$

state: powder

Zr(etac)$_4$

state : dry solid

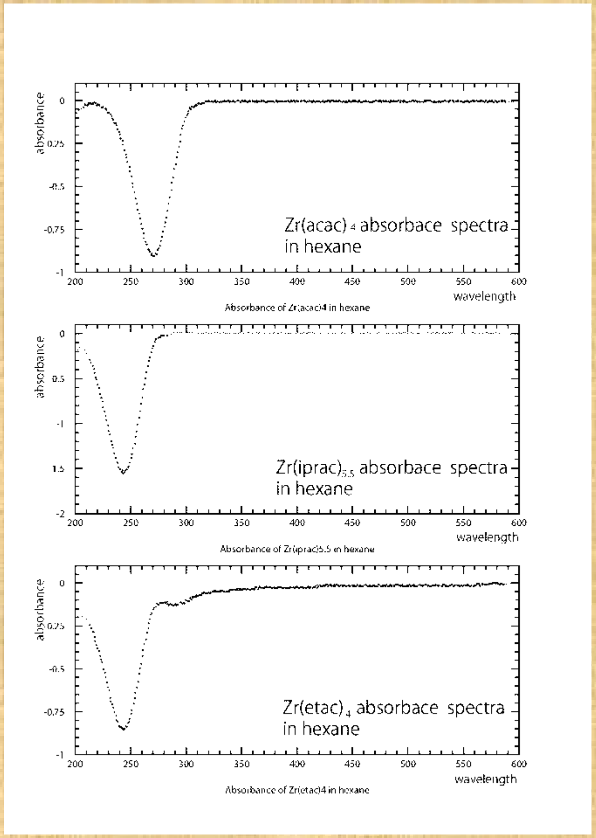


Synthesized by Prof. Takahiro Gunji (Tokyo University of Science)

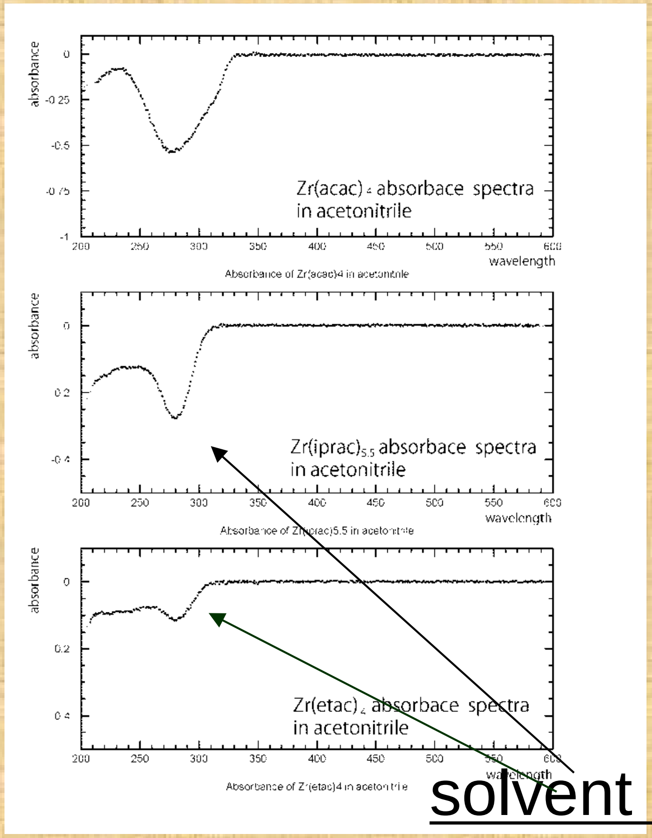
Solubility > 10 w.t.% for anisole

Absorbance spectra (Solvent effect)

Solution : Hexane



Solution : acetonitrile

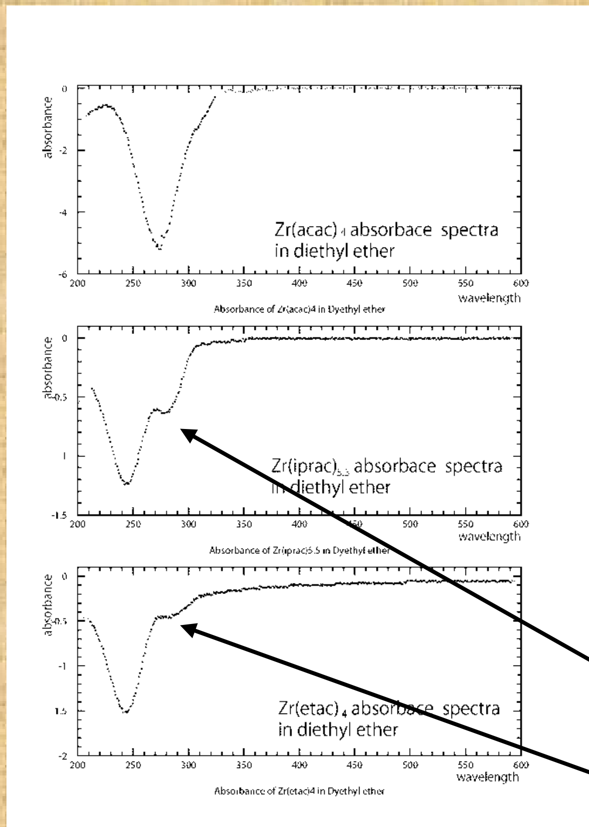


solvent effect

Absorption peak moved to shorter wavelength

Absorbance in another solvent

Solution : Diethyl Ether

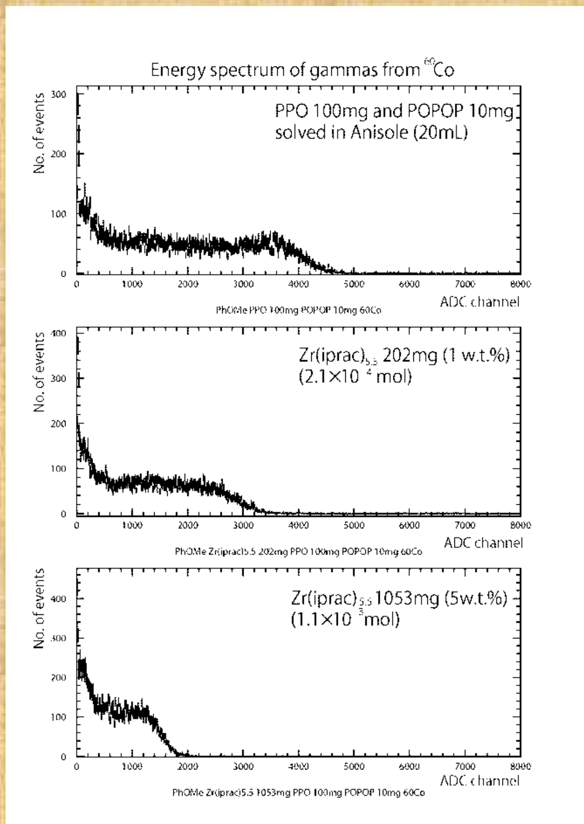


- Solvent effect could depend on the polarity (dielectric const.)
  - Acetonitrile : 37.5
  - Hexane : 1.89
  - Anisole : 4.3
- Need solution which has same polarity as anisole
  - Diethyl ether : 4.33

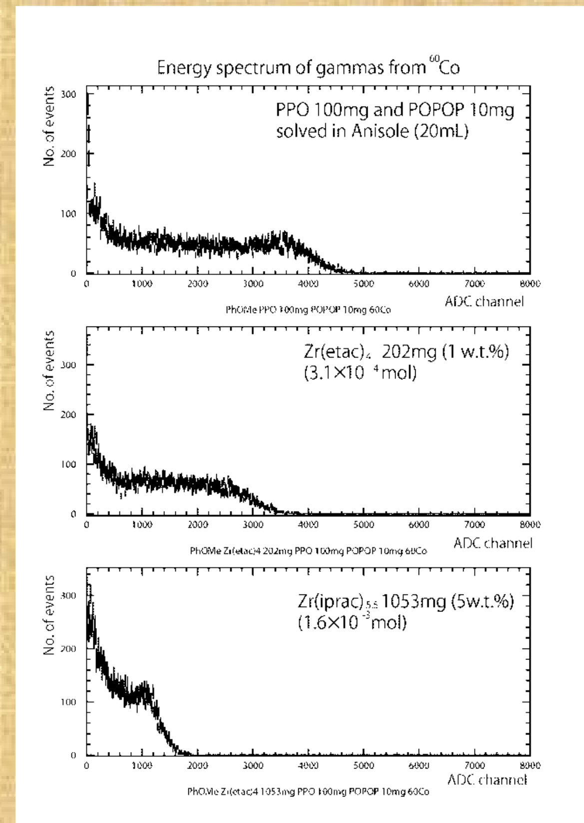
Still absorption peak remains around 270nm

Light yield of scintillation

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



Zr(etac)$_4$ in anisole



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

Requirement of scintillator solvent

- Low polarity (dielectric const.)
  - No absorption ~270nm
- Aromatic compounds
  - luminescence >270nm
- Safety for human body and environment
- Usual solvent for L.S.

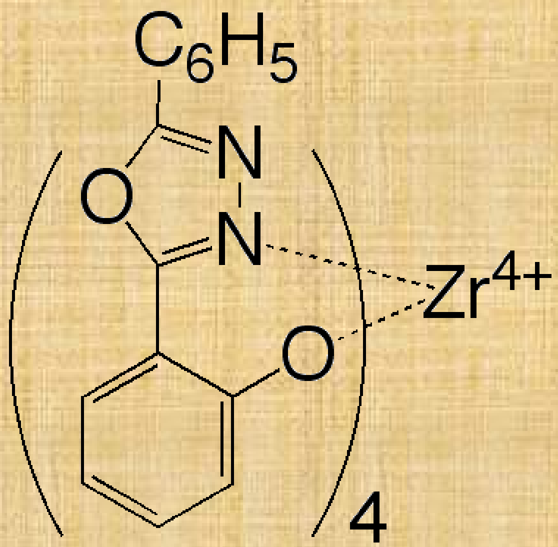
↓

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

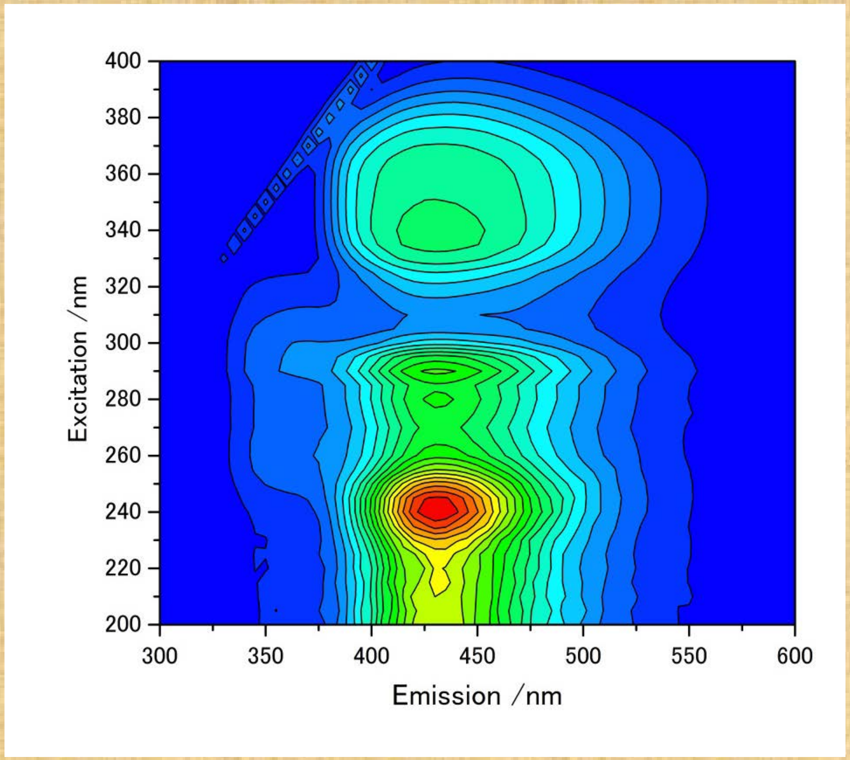
Zirconium complex with luminescence

- Zr-ODZ complex



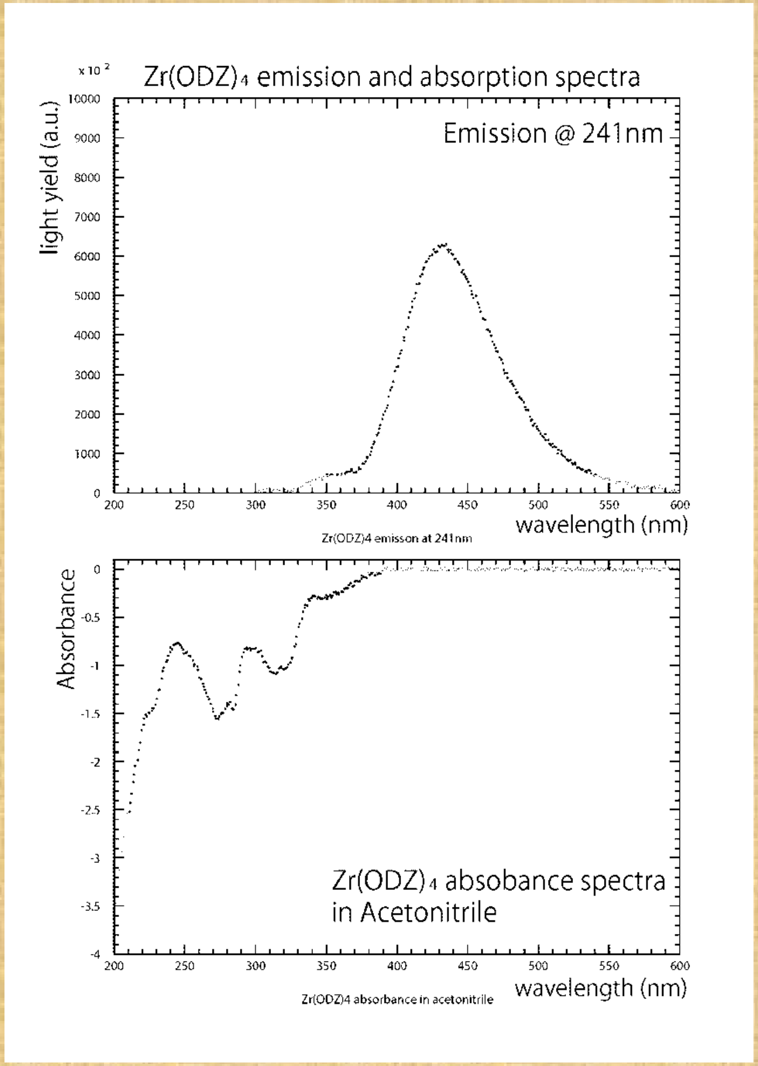
m.w. = 1040.18

- Photo luminescence



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

Emission and absorption of Zr(ODZ)$_4$



- Emission wavelength : 430nm

↓

PMT sensitive

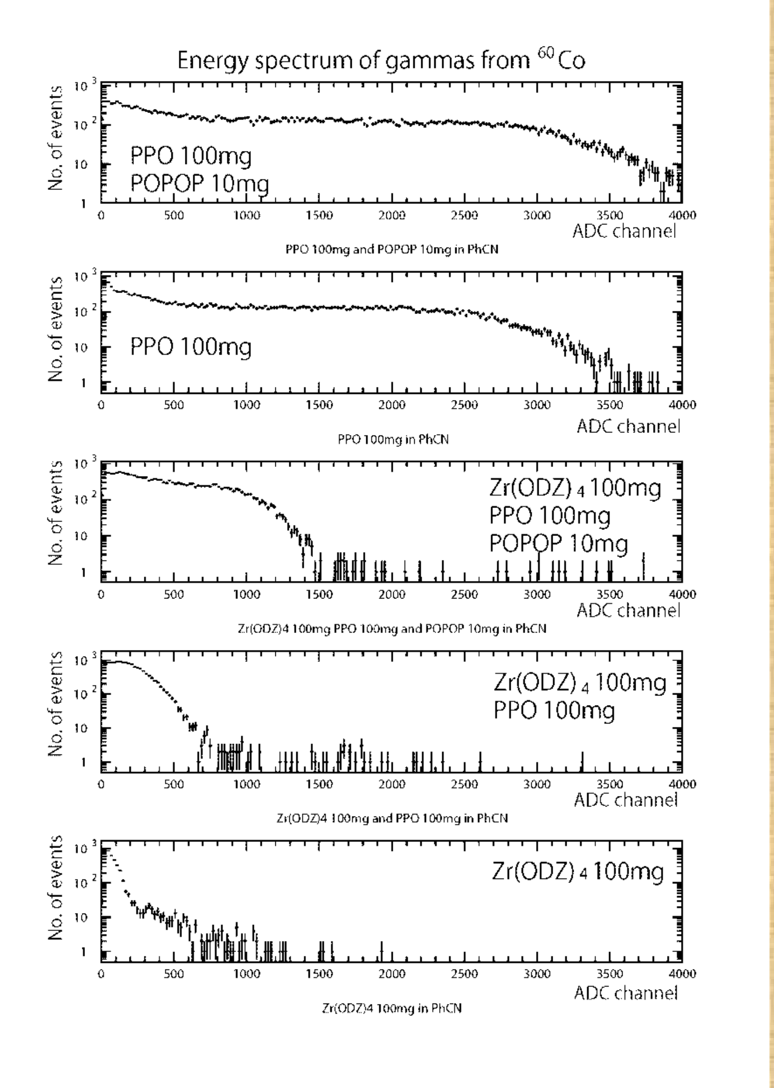
- Absorption wavelength: 270nm and 320nm

↓

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 $Zr(ODZ)_4$.
- Secondary excitation of ~340nm 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.

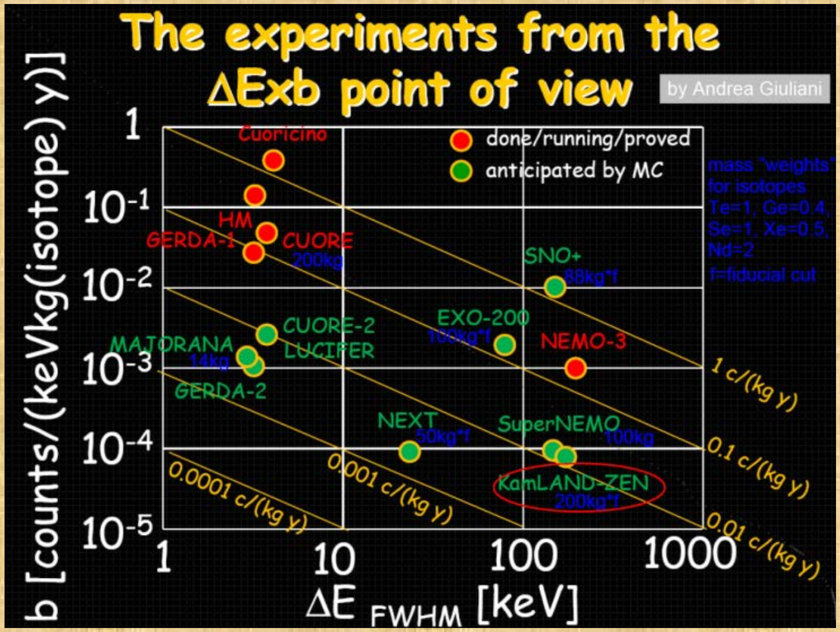
Summary

- High solubility of Zr β-keto ester in Anisole (>~10w.t.%) for ZICOS detector was achieved.
- Confirmed absorption peak moves to shorter wavelength (275nm → 245nm) by introducing substituent groups.
- Observed scintillation light yield decreased in proportion to the concentration of Zr β-keto ester due to remaining absorption @ 280nm. ⇒ Need low polarity solvent.
- Quantum yield of $Zr(ODZ)_4$ was achieved ~30%, but it was not used for scintillator due to no overlap between emission of solvent and absorption of ODZ.

BACKUP

Neutrinoless double beta decay using liquid scintillator

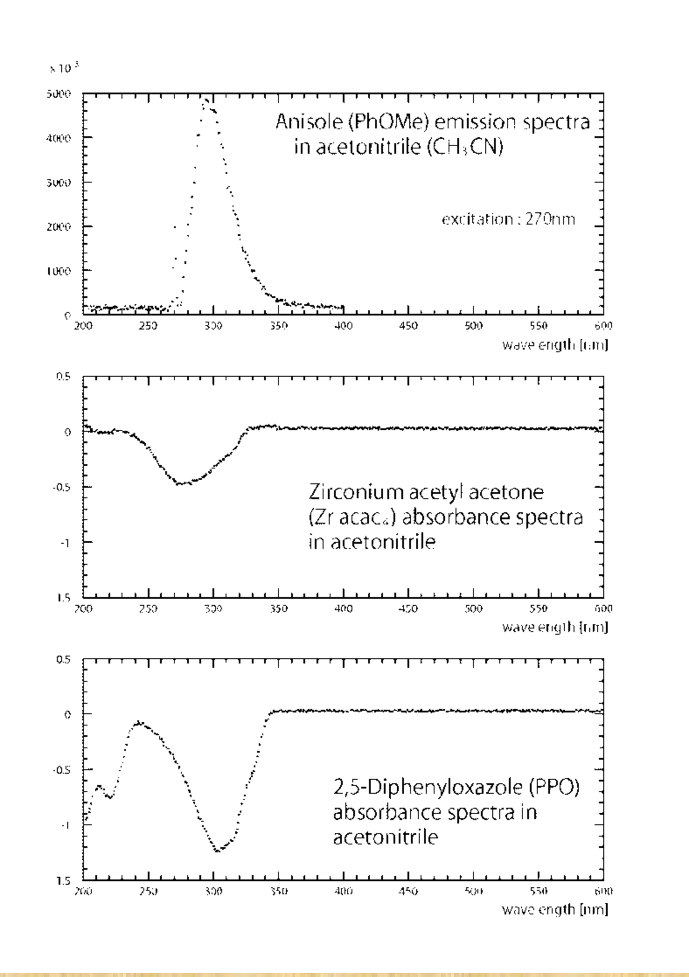
- Experimental limits for neutrino mass



- Requirement for $\langle m_\nu \rangle$:50~100meV
  - 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

What's problem : Absorption spectra of $Zr(acac)_4$



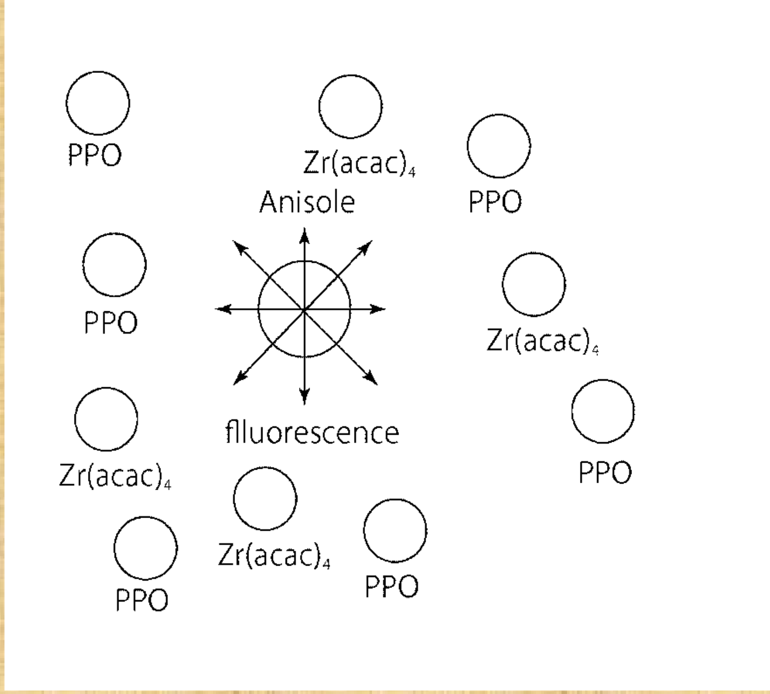
- Emission peak of anisole was observed around 295nm.
- Absorption peak of $Zr(acac)_4$ was observed around 270nm.

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

Simple expectation for quenting

- Assuming to same cross section for light

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



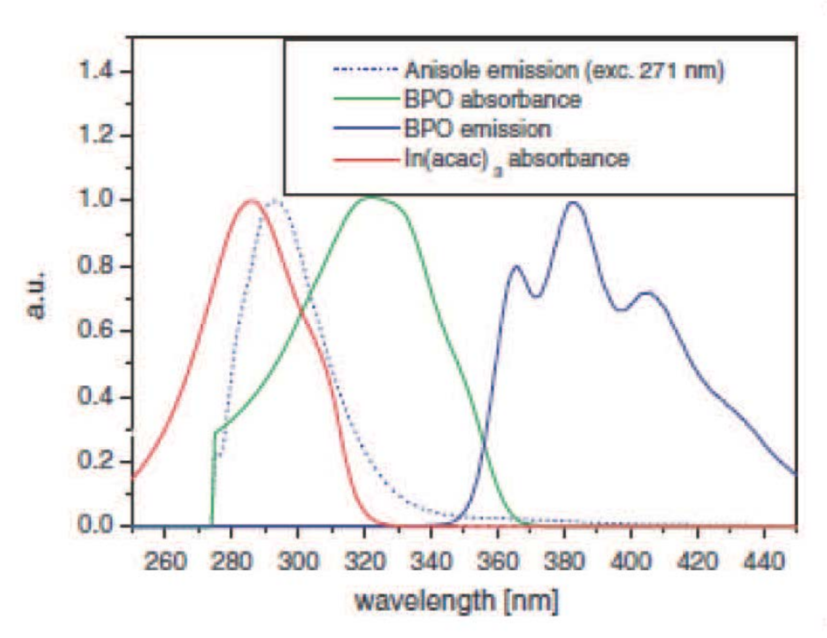
L_0 : Light yield of anisole + PPO+POPOP

N_{ppo} and N_{Zr} : No. of molecular for PPO and $Zr(acac)_4$

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

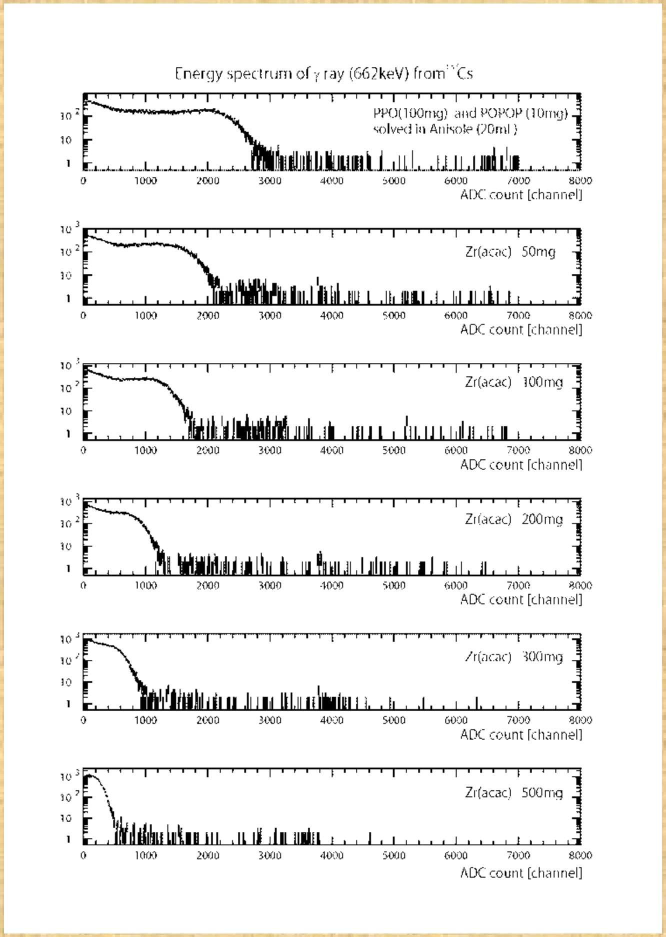
What's problem

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



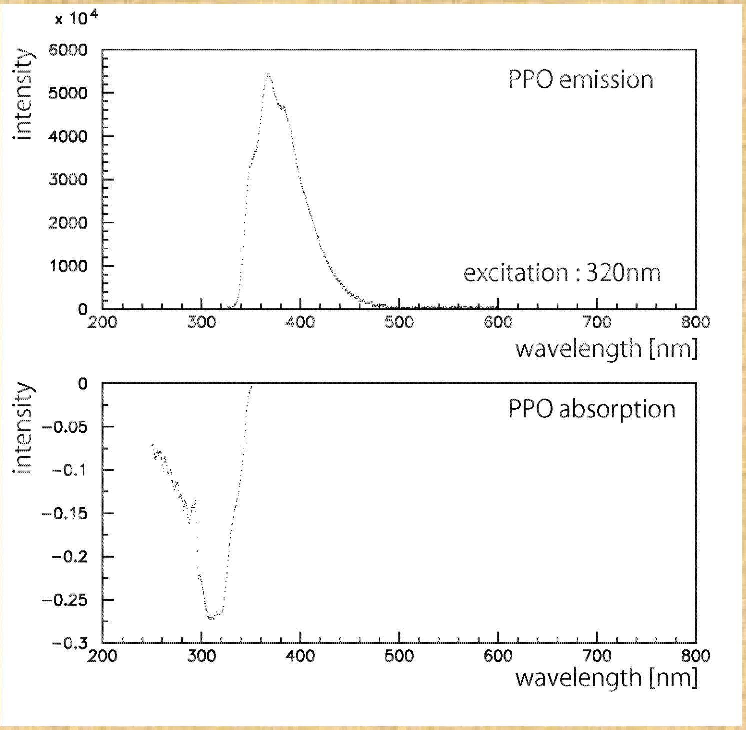
Same overlap of the emission and the absorption spectrum would be occurred even if different metal (Zr) was used.

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

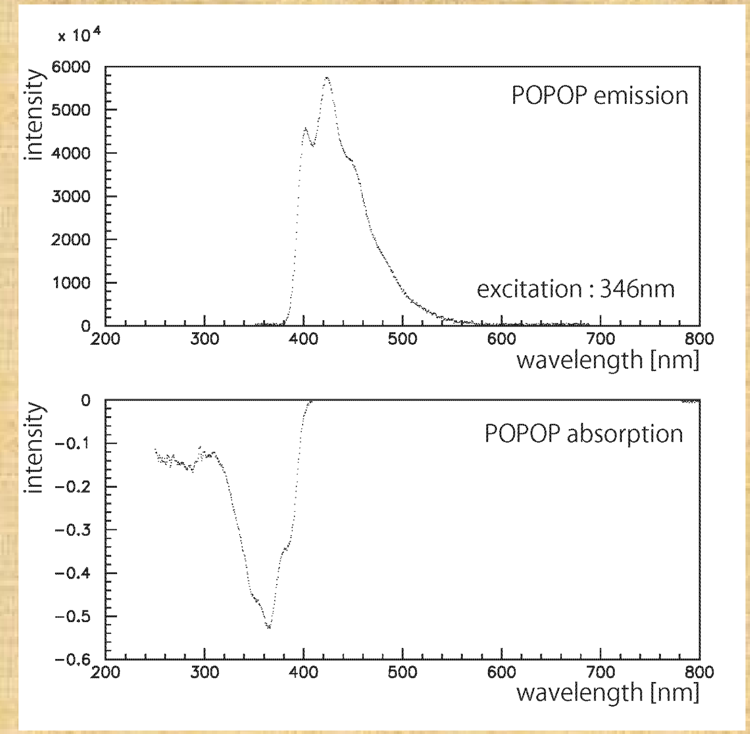
Photo Luminescence and absorption of PPO



- 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

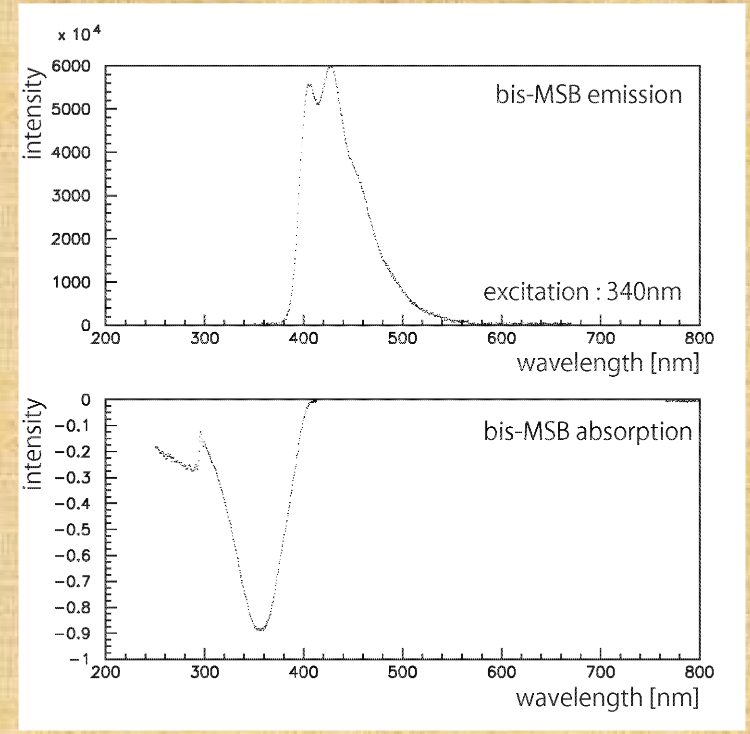
Photo Luminescence and absorption of POPOP



- 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

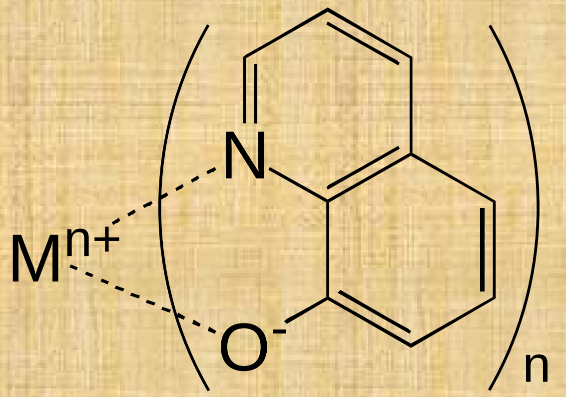


- Photo luminescence
  - Fluorescence device: HORIBA FluoroMax-4
  - Absorbance device : HITACHI U-3000
  - Solvent : Benzonitrile (PhCN)
  - Concentration : 1.0×10^{-5} mol/L

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

Response for γ–ray for tetrakis 8-quinolinolate Zr complex loaded scintillator

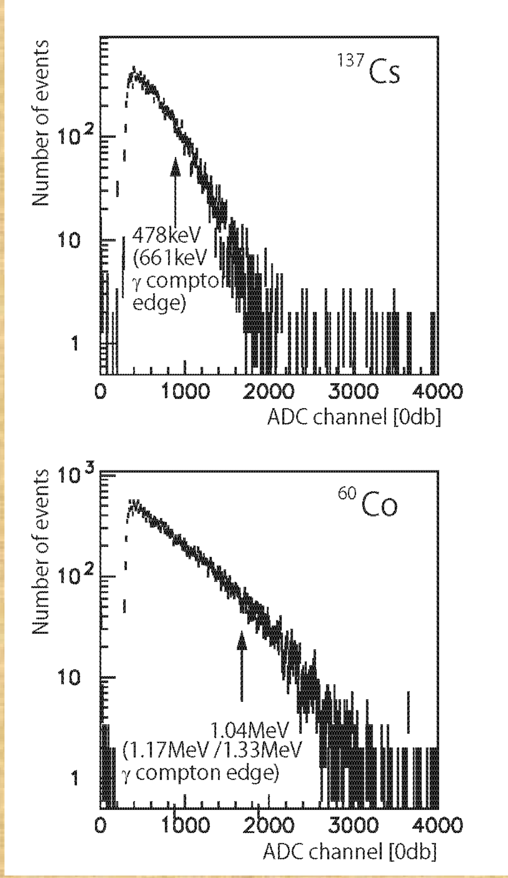
- Tetrakis (8-quinolinolate) Zirconium complex (ZrQ_4)



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%