NEWS colloquium
March 22nd, 2024

New method of high-efficiency isotope enrichment with laser

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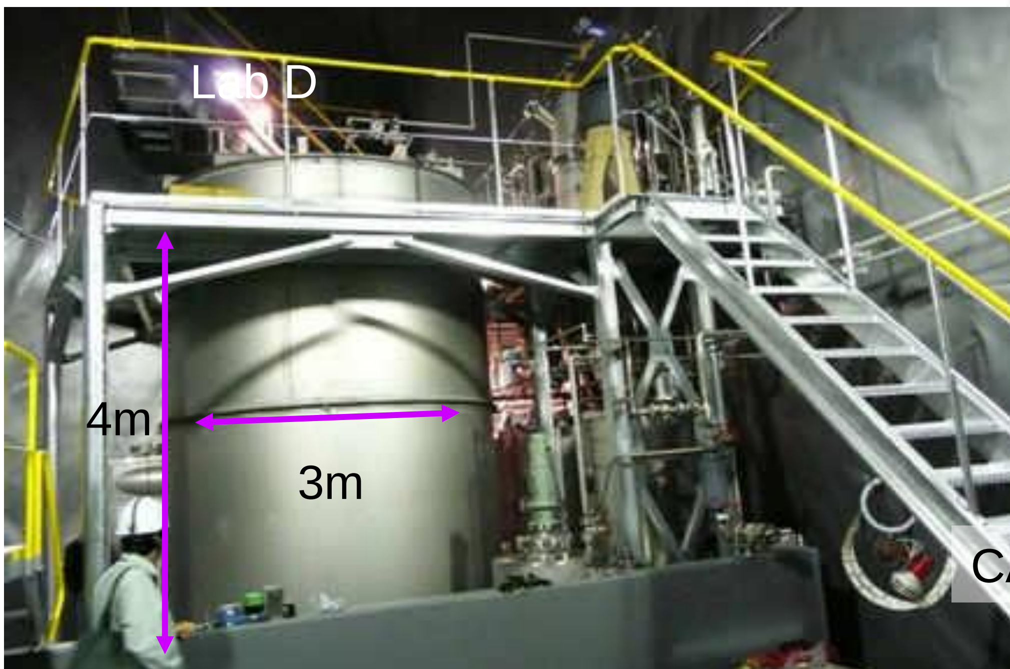
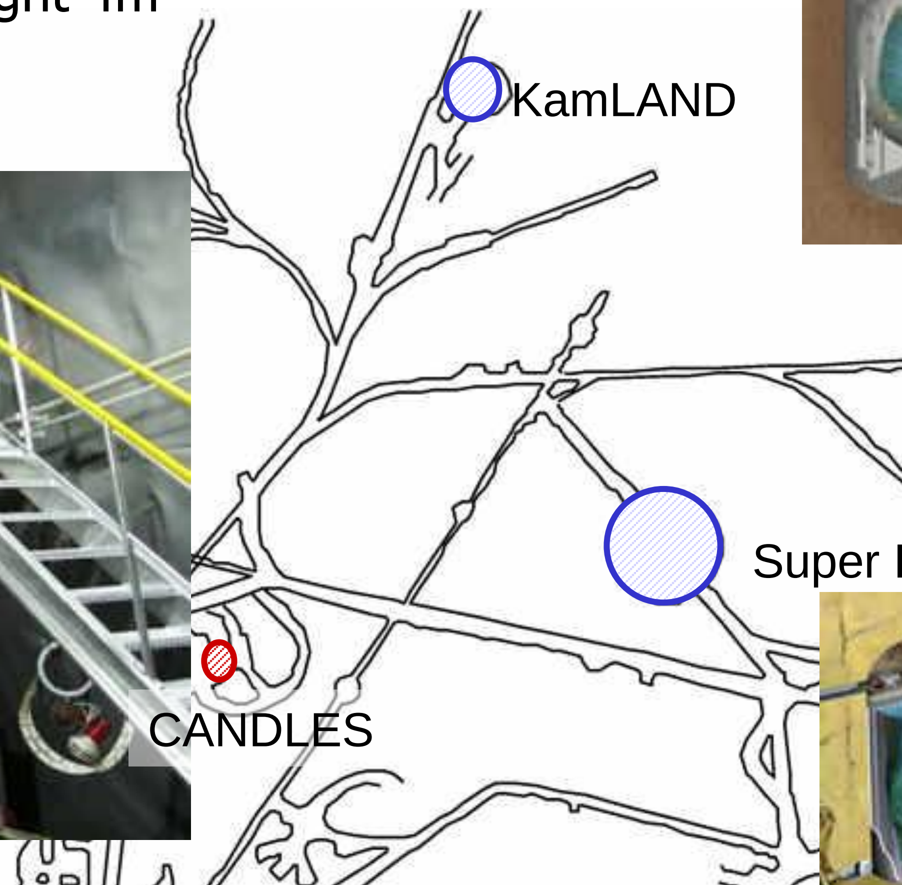
Research Center for Nuclear Physics

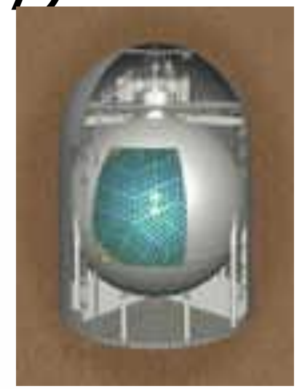
CANDLES III
Double-beta decay of ^{48}Ca

- CANDLES (Kamioka Underground Laboratory)
  - CANDLES III
    - Diameter 3m × Height 4m

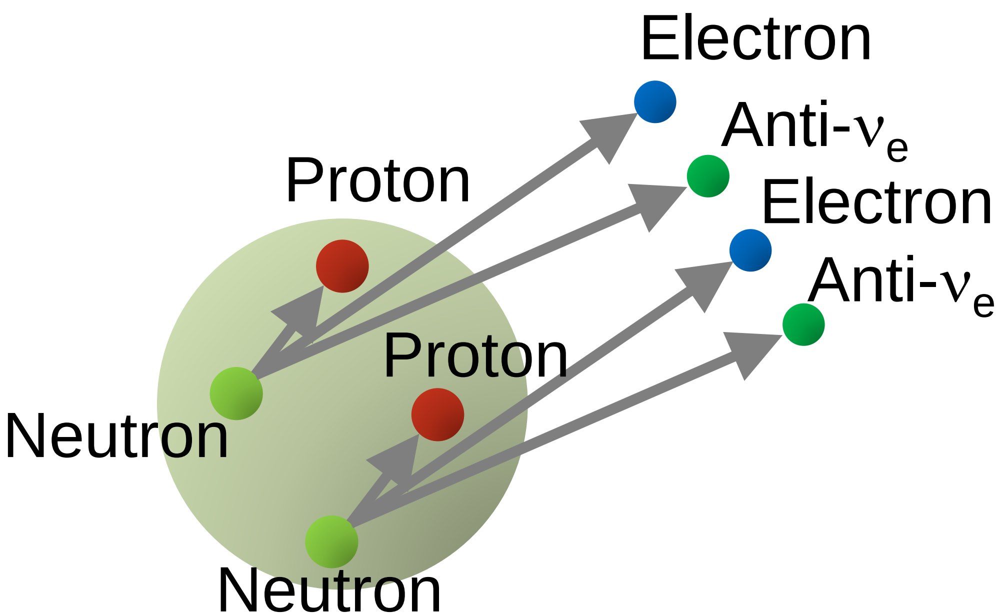


CANDLES III



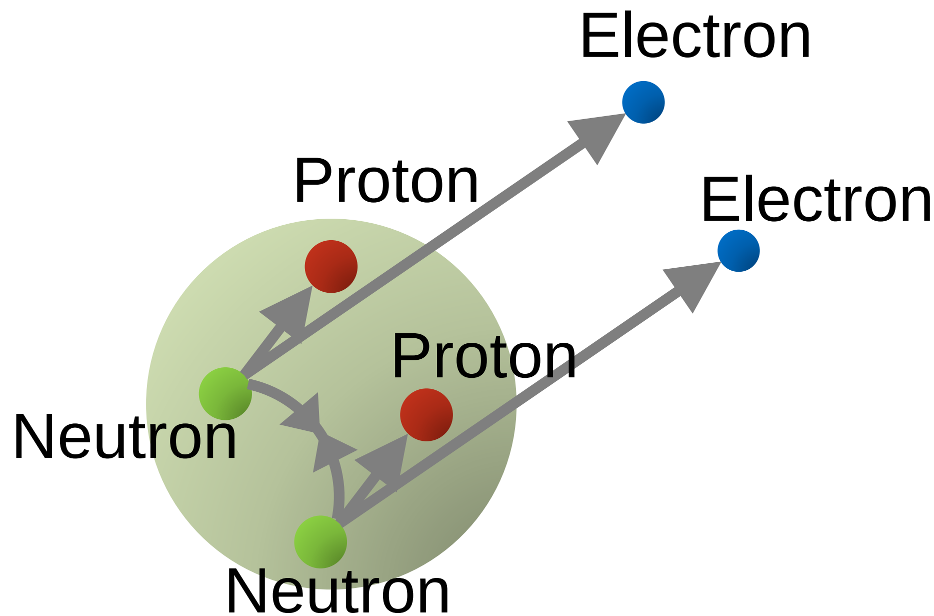


Two-neutrino mode ($2\nu\beta\beta$)



(within standard model)

Neutrino-less mode ($0\nu\beta\beta$)



(beyond standard model)

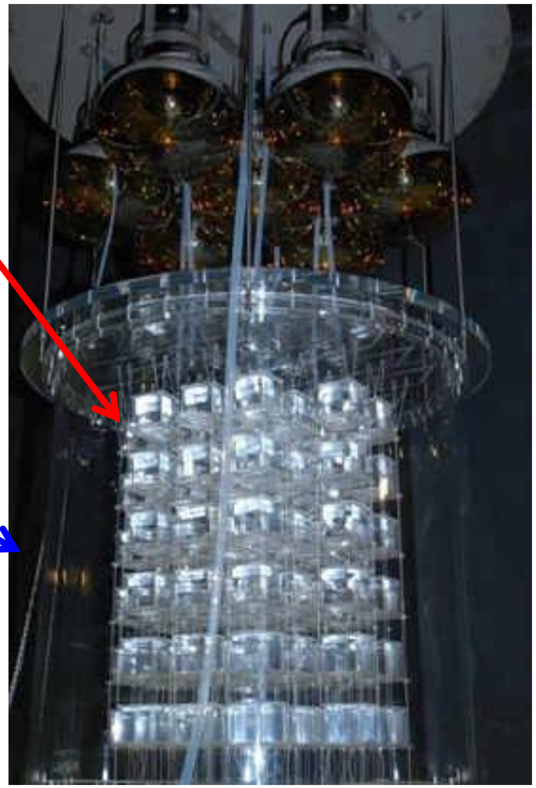
$$m_\nu > 0\ , \quad \nu \equiv \bar{\nu}$$

Majorana particle

CANDLES III

CaF_2 crystal
(305kg)

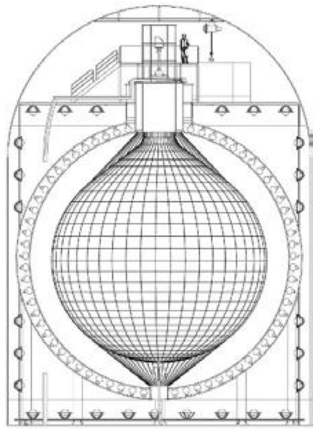
Liq. Scint.
($2m^3$)



13in. & 20in.
PM tubes

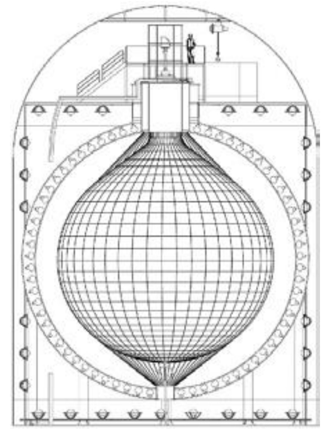
天然カルシウムに含まれる ^{48}Caは 0.187%(351g)→1トンにしたい！

KamLAND-Zen 400



until 2015, 380kg
World-best limit
$\langle m_{\beta\beta} \rangle$ < (61-165) meV

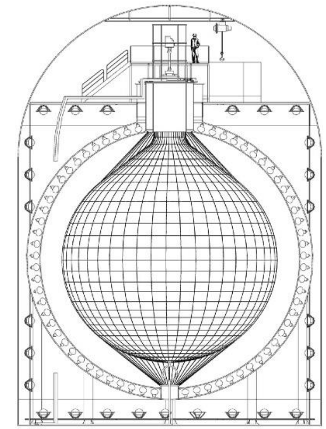
KamLAND-Zen 800



since 2019, 745kg

target 40meV

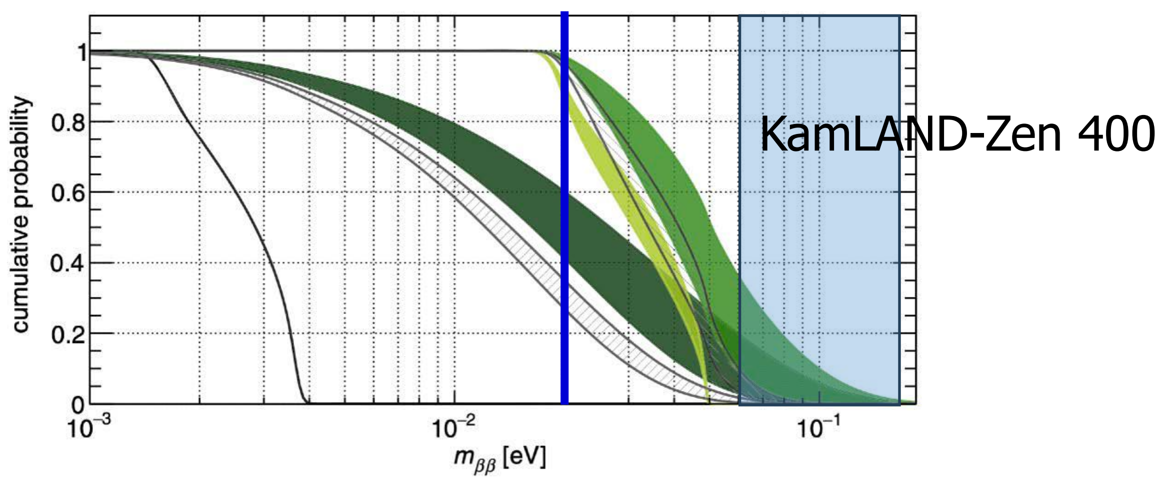
KamLAND2-Zen



future high-resolution plan

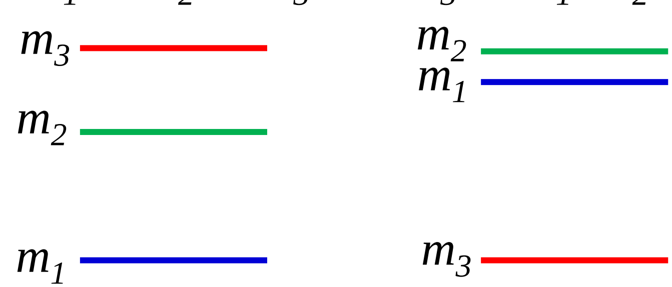
target 20meV

KamLAND2-Zen



Discovery potential;
95% of Inverted ordering
50% of Normal ordering

$m_1 << m_2 << m_3$ $\quad$ $m_3 << m_1 \sim m_2$



Normal ordering $\quad$ Inverted ordering

Notes.

(1) Normal ordering is preferred by recent data of ν-oscillation.
F. Simpson et al., JCAP 09 (2022) 006

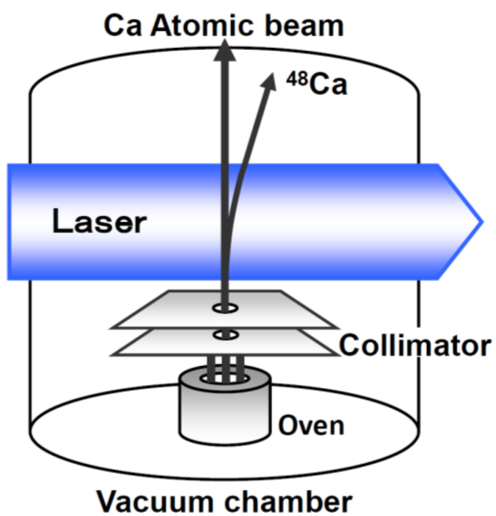
(2) Any experiment does not suggest Majorana neutrino.
It is preferred only by theoretical "naturalness".
→ Discovery potential of KamLAND2-Zen ~25% ?
It is necessary to survey down to ~1 meV.

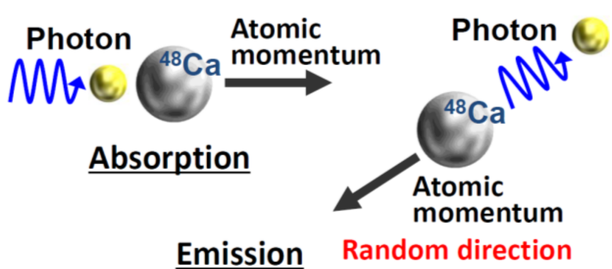
(3) Data of $T_{1/2}^{0\nu\beta\beta}$ from different nuclide are useful to separate contributions of mass term and V+A term.

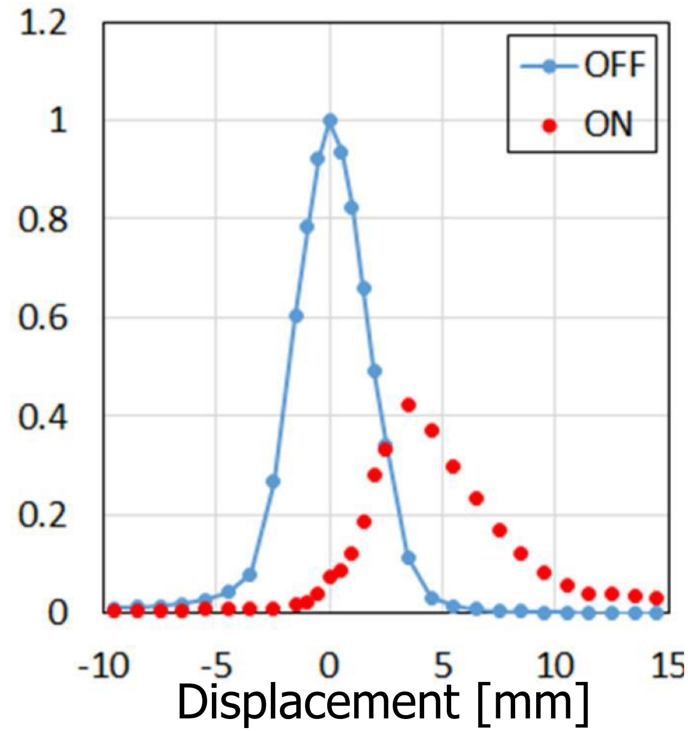
	^{48}Ca	^{76}Ge	^{82}Se	^{96}Zr	^{100}Mo	^{116}Cs	^{128}Te	^{130}Te
$R_A^{m_\nu}$	0.75	0.51	1.2	3.0	0.47	0.39	0.095	2.1
R_A^{η}	0.082	0.40	0.19	0.83	0.36	0.064	0.10	2.0
$R_A = R_A^{\eta}/R_A^{m_\nu}$	0.11	0.77	0.15	0.28	0.76	0.16	1.1	0.94

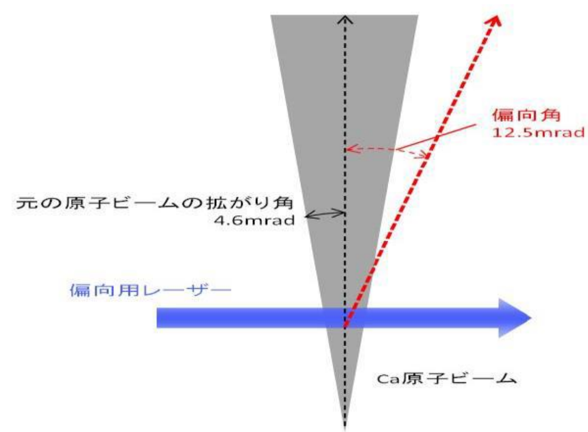
T. Fukuyama and T. Sato, JHEP04, 049 (2023)

Conventional method (→ Anawat-san)
Laser-Isotope separation (LIS)

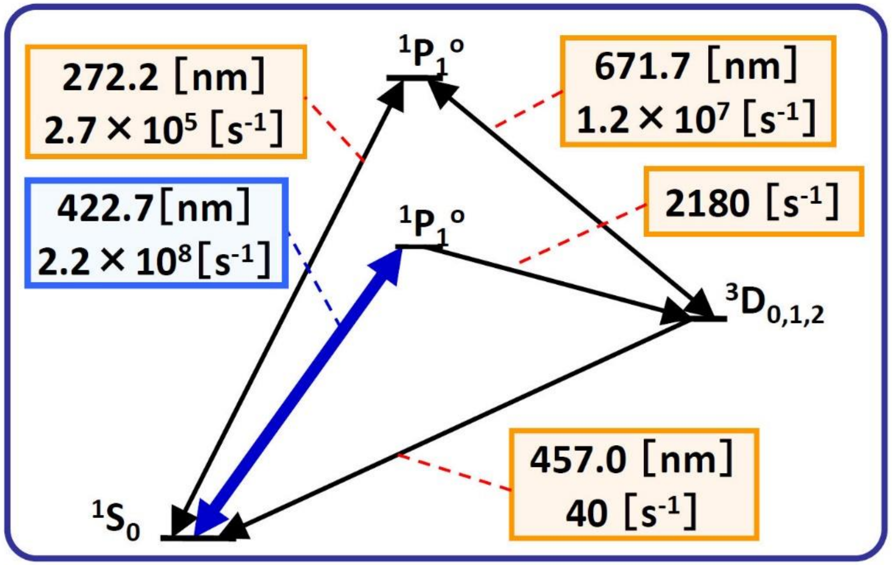








To separate ^{48}Ca atom from the atomic beam axis,
Longitudinal velocity ~800 m/s
Transvers velocity ~10 m/s → deflection angle ~12.5mrad
(~4mm shift for 300mm flight)



To get transverse velocity of 10m/s, 1000 photons of 422.7nm (2.93eV) should be absorbed by one atom
→ To produce 1ton,

$$6\times10^{23}\times2.93\times1000\times\frac{10^6}{48}=3.67\times10^{31}\ [\mathrm{eV}]$$

$$=5.88\times10^{12}\ [\mathrm{J}]\sim\ 6\ [\mathrm{TJ}]$$

is necessary.

Current target quantity is 1 mol/year.

Photon cost

(based on Niki-sensei's estimation)

6TJ is needed to produce 1 ton

Cf. Today's beam power of J-PARC 3GeV booster is 879kW.

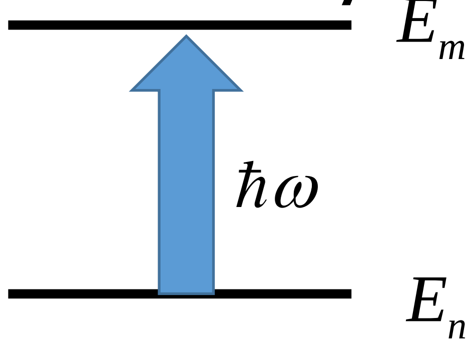
→ Laser power 190kW to produce 1 ton in one year

Kinetic energy of 1 ton mass with transverse velocity of 10m/s is
$1/2 \times 1000\mathrm{kg} \times (10\mathrm{m/s})^2 = 50\mathrm{kJ}$ --- $< 10^{-5} \times 6\mathrm{TJ}$

★ Most of the laser power is lost in spontaneous emission.
★ To recycle deexcitation photons, it is needed to make deexcitation with not spontaneous emission but stimulated emission.
★ To make stimulated emission, guide beam is necessary.
★ To shoot the excited atoms with a guide beam, it is necessary to make deexcitation at the known position and time.

⇒ **Rabi oscillation**

Two-level system



$$i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t}=H\psi(\mathbf{r},t)$$

$$H=H_0+H_{\text{int}}$$

$$H_0\varphi_n(\mathbf{r})=E_n\varphi_n(\mathbf{r})$$

$$\psi=\sum_n b_n(t)\exp(-iE_nt)\cdot\varphi_n(\mathbf{r})$$

$$\sum_n\left|b_n(t)\right|^2=1$$

$$E_{ex}=E_m-E_n=\hbar\omega_{mn}$$

$$i\frac{\partial}{\partial t}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}=\begin{pmatrix}E_n & \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t)\\ \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t) & E_m\end{pmatrix}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}$$

Optical Bloch eq.

$\mathbf{E}(t)=\mathbf{E}_0\sin(\omega t)$; laser electric field

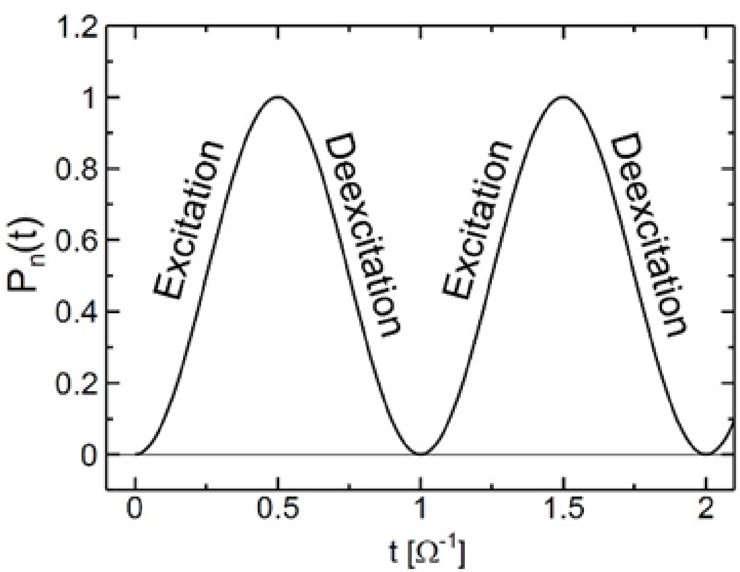
$\boldsymbol{\mu}_{mn}$; transition dipole moment between m and n

$$i\frac{\partial}{\partial t}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}=\begin{pmatrix}E_n & \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t)\\ \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t) & E_m\end{pmatrix}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}$$

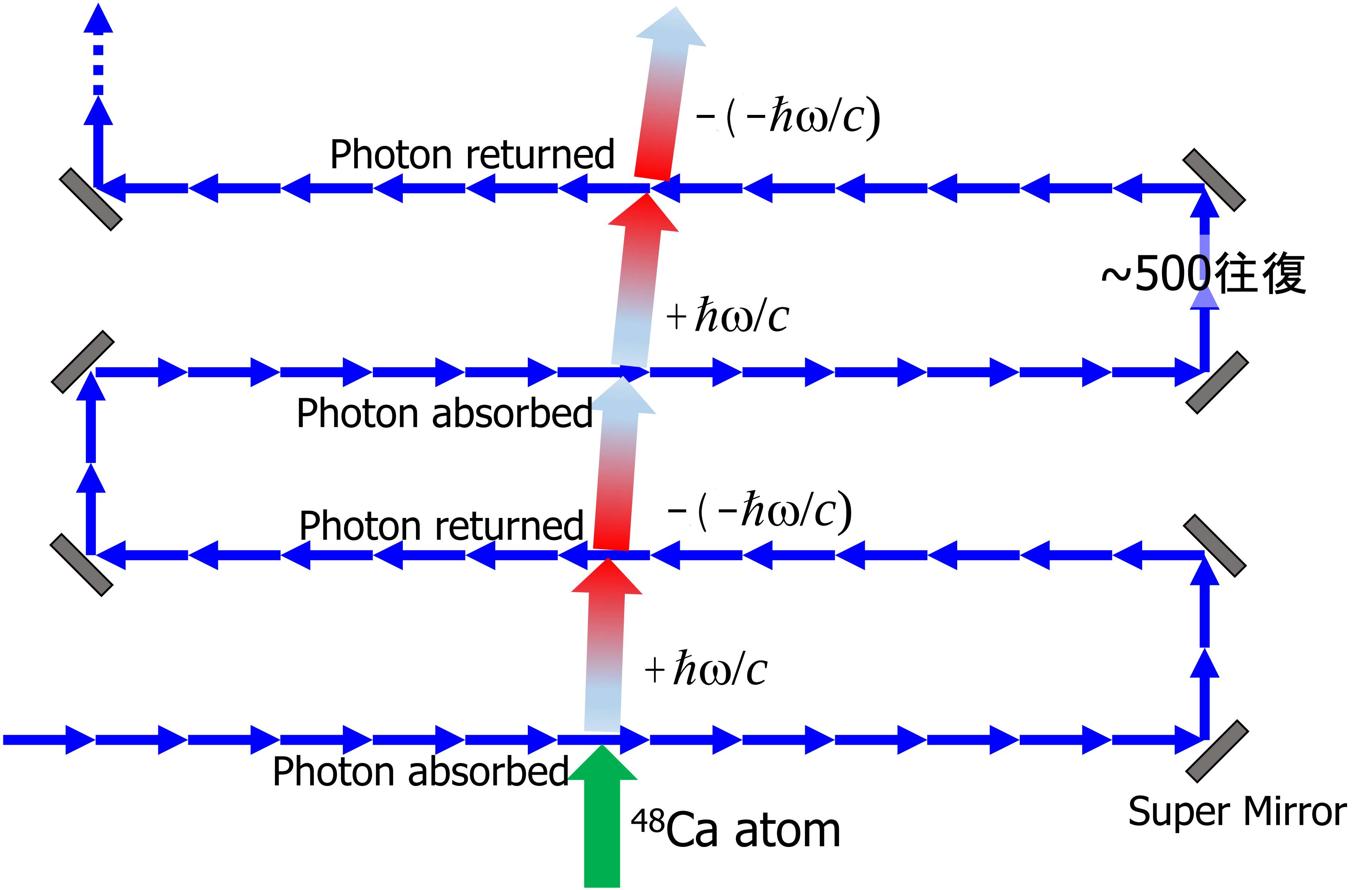
For $\psi_n(0)=1,\ \psi_m(0)=0,$

$$\left|\psi_m(t)\right|^2=\frac{\Omega^2}{\delta^2+\Omega^2}\cdot\sin^2\left(\frac{\sqrt{\delta^2+\Omega^2}}{2}t\right)\ ,\quad \text{where } \delta\equiv\omega-\omega_{mn}\ ,\ \ \Omega\equiv\frac{\boldsymbol{\mu}_{mn}\cdot\mathbf{E}_0}{\hbar}$$

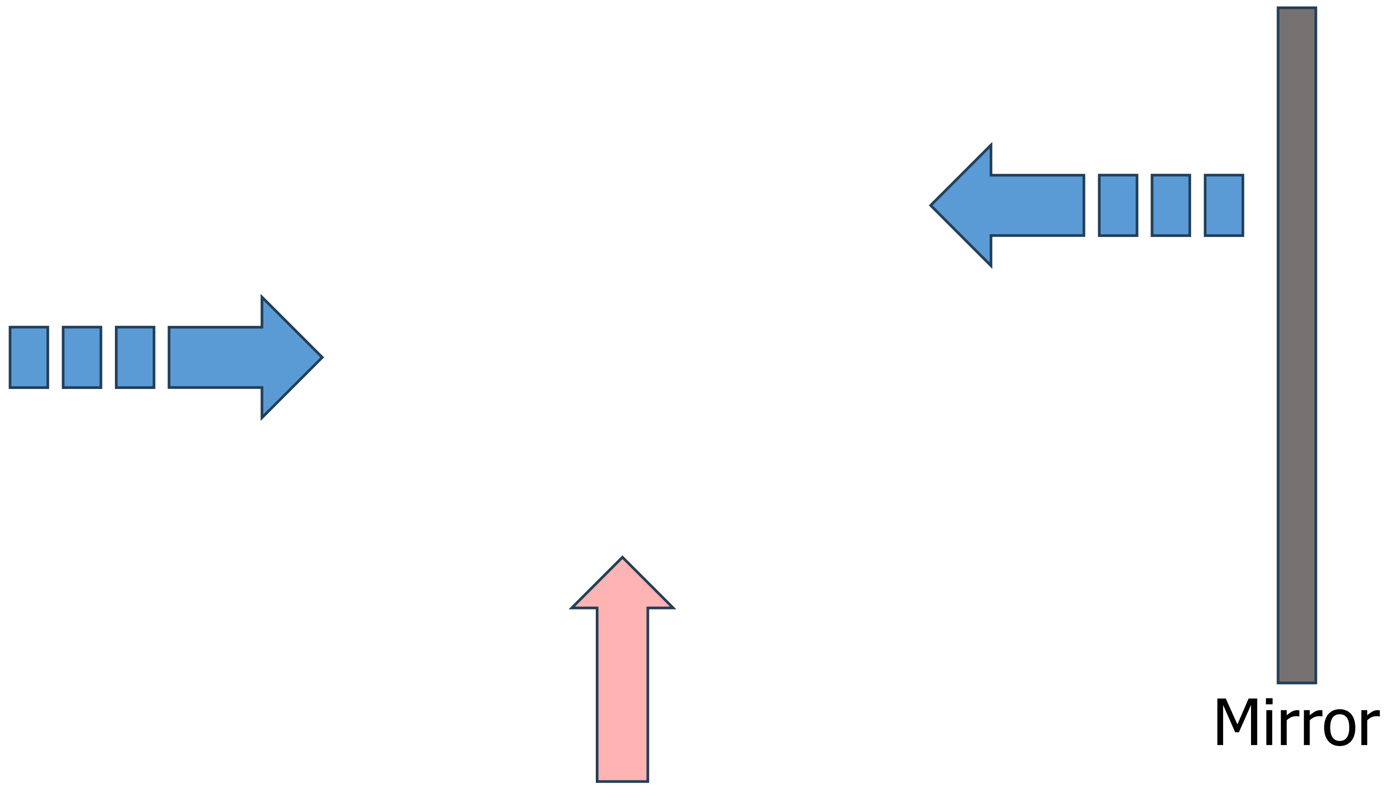
If $\delta=0$ i.e. $\omega=\omega_{mn}$, $\quad \left|\psi_m(t)\right|^2=\sin^2\left(\frac{\Omega}{2}t\right)=\frac{1-\cos\Omega t}{2}$

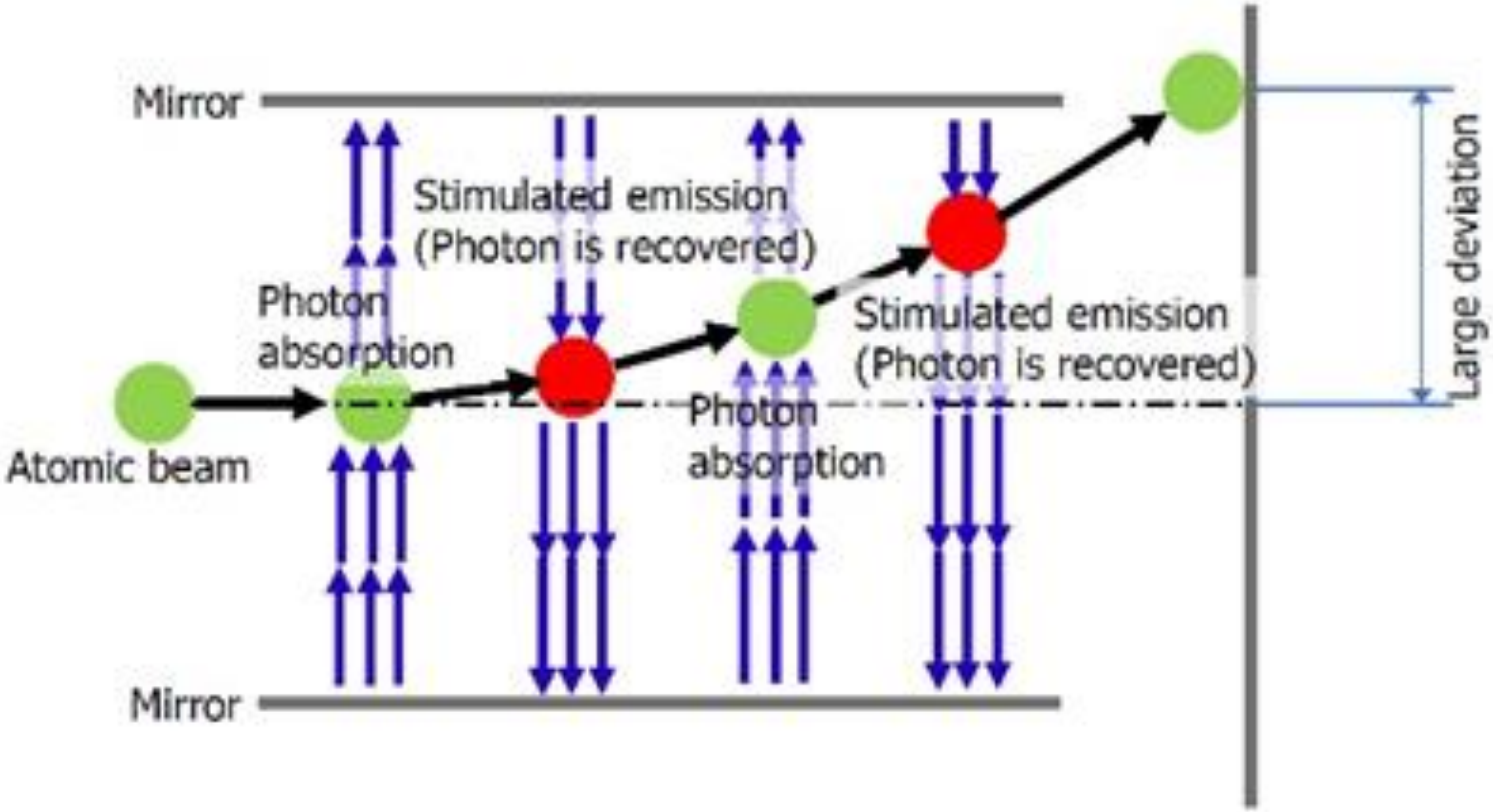


"Rabi oscillation"
Photon absorption and emission occur at definite timing.



Photons emitted by stimulated emission should carry the same energy, momentum and polarization with guide beam.

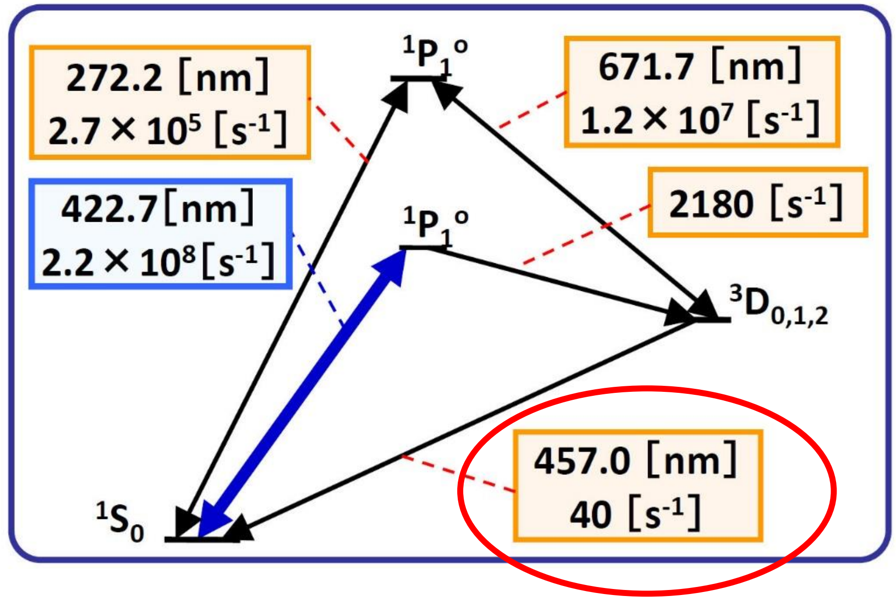

Mirror



Mirror
Stimulated emission
(Photon is recovered)
Photon
absorption
Atomic beam
Stimulated emission
(Photon is recovered)
Photon
absorption
Large deviation
Mirror

Required condition for Rabi frequency Ω :

(1) Stimulated emission rate should be much higher than spontaneous emission rate (~40Hz).



(2) Harmonics due to pulsing should not overlap with resonance frequency of neighboring isotope (Δf ~800MHz).

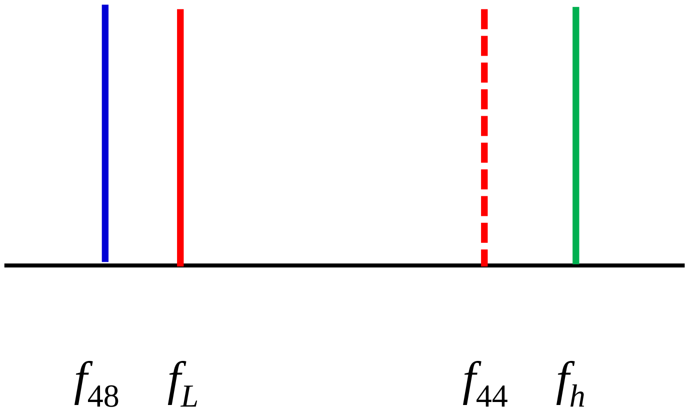
(3) Amplitude $\frac{\Omega^2}{\delta^2+\Omega^2}$ should be close to unity for detuning δ (~80MHz) due to Doppler shift.

Effect of harmonics due to pulsing

Fourier series of square wave

$$f(t) = A\sin(2\pi ft)\cdot H(f_H, t)$$
$$= A\sin(2\pi ft)\cdot\sum_{n=1}^{\infty}\frac{4}{\pi}\frac{1}{2n-1}\sin(2\pi(2n-1)f_H t)$$
$$= A\sin(2\pi ft)\cdot\frac{4}{\pi}\left\{\sin(2\pi f_H t)+\frac{1}{3}\sin(6\pi f_H t)+\frac{1}{5}\sin(10\pi f_H t)+\ldots\right\}$$
$$= \left[f \pm f_H, f \pm 3f_H, f \pm 5f_H, \cdots\right]$$

Sideband may excite the neighboring isotope; Δf=800MHz



$$f_{48} - f_{44} = 800 \text{ [MHz]}$$

$$f_L - f_h = \Omega$$

$$I_{44} = \frac{\Omega^2}{\left(f_{44} - f_h\right)^2 + \Omega^2} \cdot \left(\frac{I_h}{I_L}\right) = \frac{\Omega^2}{\left(\Omega - 800\right)^2 + \Omega^2} \cdot \frac{1}{9} < 0.01 \text{ --- (1)}$$

$$I_{44} = \frac{\Omega^2}{\left(f_{44} - f_L\right)^2 + \Omega^2} = \frac{\Omega^2}{\delta^2 + \Omega^2} > 0.99 \text{ --- (2)}$$

From (1); $\Omega^2 < 0.09\left(\left(\Omega - 800\right)^2 + \Omega^2\right) \Rightarrow (1\text{-}0.18)\Omega^2 < 0.09 \cdot 800^2 - 144\Omega$

$$\Rightarrow 82\Omega^2 - 9 \cdot 800^2 + 14400\Omega < 0 \Rightarrow \left(\Omega + \frac{7200}{82}\right)^2 < \left(\frac{9 \cdot 800^2}{82} + \frac{7200^2}{82^2}\right)$$

$$\frac{-\sqrt{82 \cdot 9 \cdot 800^2 + 7200^2}}{82} < \Omega + \frac{7200}{82} < \frac{+\sqrt{82 \cdot 9 \cdot 800^2 + 7200^2}}{82}$$

$$\Rightarrow \frac{-7200 - \sqrt{82 \cdot 9 \cdot 800^2 + 7200^2}}{82} < \Omega < \frac{-7200 + \sqrt{82 \cdot 9 \cdot 800^2 + 7200^2}}{82}$$

$$\frac{-7200 - 22894.5}{82} < \Omega < \frac{-7200 + 22894.5}{82} \Rightarrow -367 \text{ [MHz]} < \Omega < 191.4 \text{ [MHz]}$$

From (2); $\Omega^2 > 0.99\left(\delta^2 + \Omega^2\right) \Rightarrow 0.1\Omega > \delta \Rightarrow \Omega > 360$ [MHz]

- - - (1) and (2) are not satisfied simultaneously. $\Rightarrow$ Higher priority to Eq. (1)

If $\Omega = 190$ [MHz]

$$I_{44} = \frac{500^2}{300^2 + 500^2} \cdot \left(\frac{1}{3}\right)^2 = 0.0098$$

$$I_{48} = \frac{500^2}{36^2 + 500^2} = 0.965$$

Ω=190 MHz → Laser power of 38.8kW is needed.
Assuming finesse of cavity as 10^5 → 0.388W (peak)
~ 200mW (mean)

$$i\frac{\partial}{\partial t}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}=\begin{pmatrix}E_n & \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t)\\ \boldsymbol{\mu}_{mn}\cdot\mathbf{E}(t) & E_m\end{pmatrix}\begin{pmatrix}\psi_n(t)\\ \psi_m(t)\end{pmatrix}$$

For $\psi_n(0)=1,\ \psi_m(0)=0,$

$$\left|\psi_m(t)\right|^2=\frac{\Omega^2}{\delta^2+\Omega^2}\cdot\sin^2\left(\frac{\sqrt{\delta^2+\Omega^2}}{2}t\right)\ ,\quad \text{where } \delta\equiv\omega-\omega_{mn}\ ,\ \Omega\equiv\frac{\boldsymbol{\mu}_{mn}\cdot\mathbf{E}_0}{\hbar}$$

If $\delta=0$ i.e. $\omega=\omega_{mn}$, $\left|\psi_m(t)\right|^2=\sin^2\left(\frac{\Omega}{2}t\right)=\frac{1-\cos\Omega t}{2}$

Transition dipole moment $\boldsymbol{\mu}_{mn}$ is determined from lifetime of spontaneous emission

$$A = \frac{\omega_0^3 \mu_{mn}^2}{3\pi\varepsilon_0 \hbar c^3} = \frac{(2\pi)^3 f_0^3 \mu_{mn}^2}{3\pi\varepsilon_0 \hbar c^3} = \frac{8\pi^2}{3\varepsilon_0 \hbar \lambda^3}\mu_{mn}^2$$

$$\varepsilon_0 = 8.854 \times 10^{-12}\ [\mathrm{F/m}] = 8.854 \times 10^{-12}\ [\mathrm{C/V \cdot m}]$$

$$\mu_{mn} = \sqrt{\frac{3\varepsilon_0 \hbar \lambda^3 A}{8\pi^2}} \qquad \hbar = 1.055 \times 10^{-34}\ [\mathrm{J \cdot s}] = 1.055 \times 10^{-34}\ [\mathrm{C \cdot V \cdot s}]$$

$$= \sqrt{\frac{3 \times 8.854 \times 10^{-12} \times 1.055 \times 10^{-34} \times (4.23 \times 10^{-7})^3 \times 40}{8\pi^2}} = 1.16 \times 10^{-32}\ [\mathrm{C \cdot m}]$$

$$\Omega \equiv \frac{\boldsymbol{\mu}_{mn} \cdot \mathbf{E}_0}{\hbar} = 190\ [\mathrm{MHz}] \quad \Rightarrow \quad |\mathbf{E}_0| = \frac{\Omega}{\mu_{mn}}\hbar$$

$$\Omega = 190\ [\mathrm{MHz}],\ \mu_{mn} \sim 1.16 \times 10^{-32}\ [\mathrm{C \cdot m}],\ \hbar = 1.055 \times 10^{-34}\ [\mathrm{J \cdot s}]$$

$$\Rightarrow \quad |\mathbf{E}_0| \sim 1.73\ [\mathrm{MV/m}]$$

Energy density; $p = \varepsilon|\mathbf{E}_0|^2 = 8.854 \times 10^{-12}\ [\mathrm{F/m}] \times 2.99 \times 10^{12}\ [\mathrm{V^2/m^2}]$
$= 26.5\ [\mathrm{J/m^3}]$

$$p = \frac{P\ [\mathrm{W}]}{cA} = \frac{P\ [\mathrm{J/s}]}{3 \times 10^8\ [\mathrm{m/s}] \times \pi \times (1.25 \times 10^{-3}\ [\mathrm{m}])^2} \cong \frac{P}{1472.6}\ [\mathrm{J/m^3}]$$

$$\Rightarrow \quad P = 1472.6p\ [\mathrm{W}] = 38.8\ [\mathrm{kW}] \quad (\text{ for one Ca atom. })$$

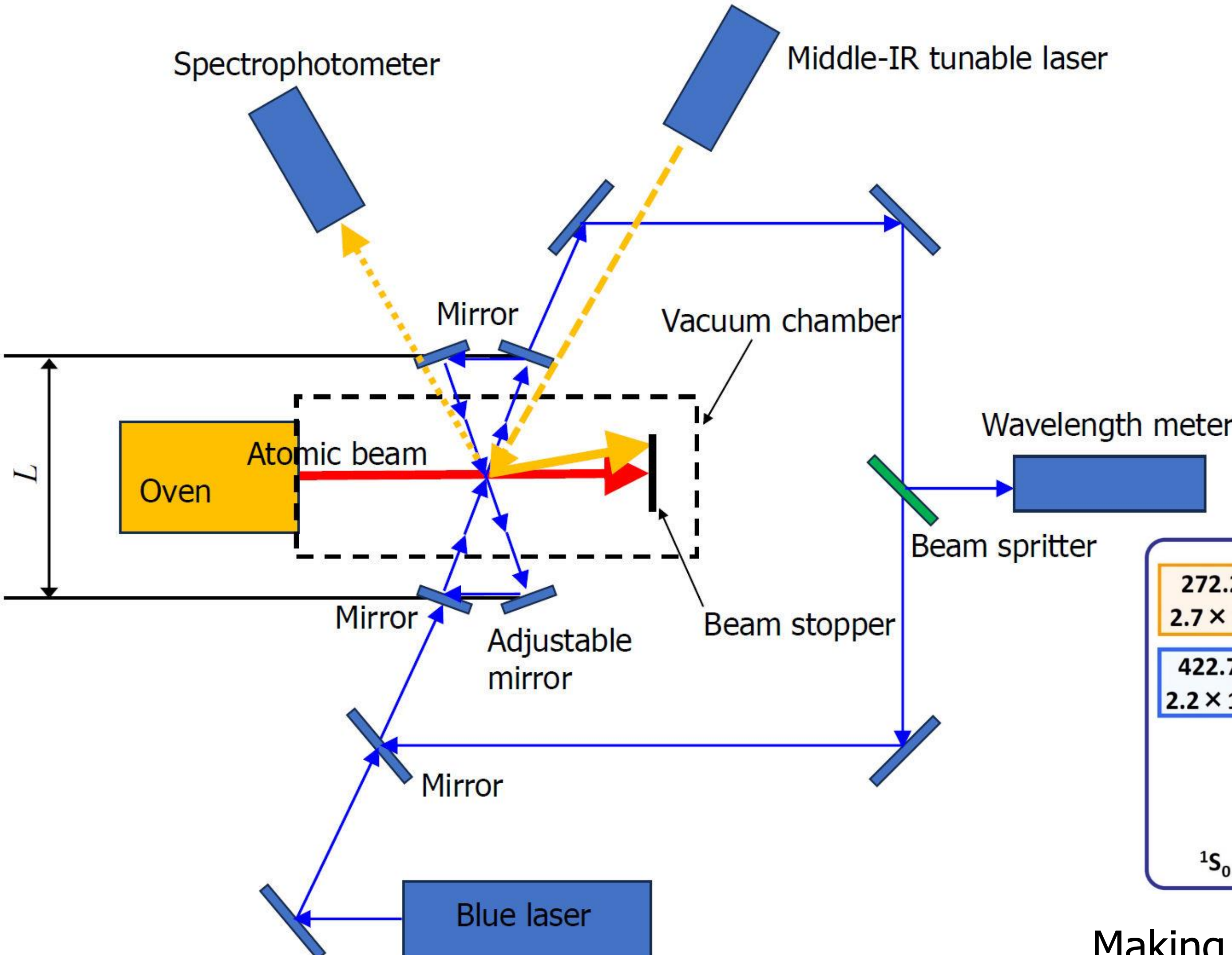
$P = 38.8\ [\mathrm{kW}]$ (for one Ca atom.)

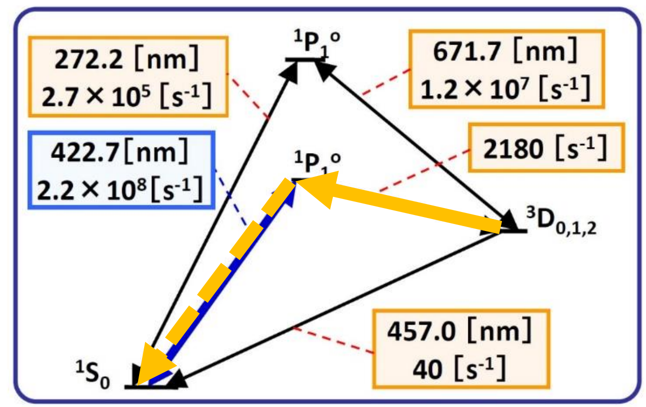
$$\Rightarrow \quad I_{photon} = \frac{P}{h\nu} = \frac{38.8 \times 10^3}{6.626 \times 10^{-34} \times \frac{3 \times 10^8}{4.5 \times 10^{-7}}} = 8.784 \times 10^{22}\ [\mathrm{photon/s}]$$

--- sufficient to establish stimulated absorption/emission

Since the photons emitted by deexcitation ware recovered, one can use optical cavity to obtain high-density photons. If the “finesse” (Q-value of the cavity) is as high as 10^5, required laser power is 388mW.

To produce 1 ton in one year, production rate should be $2.4\mathrm{mol/h} = 4 \times 10^{20}\ {}^{48}Ca$ atoms/s
If one use 30 units in the same time, 1.3×10^{19} atoms/s/unit $<< 8.784 \times 10^{22}$ photons/s





Making 3D → $1P_1^0$ transition with 5.632 μm laser and observe 422.7 nm deexcitation photon to detect Rabi oscillation.

Intensity of atomic beam

$$\Phi = n_0 \cdot \left(\frac{\gamma k_B T_0}{m} \right)^{1/2} \cdot \left(\frac{2}{\gamma + 1} \right)^{\frac{\gamma+1}{2(\gamma-1)}} \quad , \ \gamma = \frac{5}{3} \ \text{for mono-atomic molecule}$$

$$= n_0 \cdot \left(\frac{5 k_B T_0}{3m} \right)^{1/2} \cdot \left(\frac{3}{4} \right)^2 = \frac{1\ [\text{mol}]}{22400\ [\text{cm}^3]} \cdot \sqrt{\frac{5 \times 75\ [\text{meV}]}{3 \times 48 \times \frac{938.3\ [\text{MeV}]}{1.0073}}} \cdot \frac{9}{16}$$

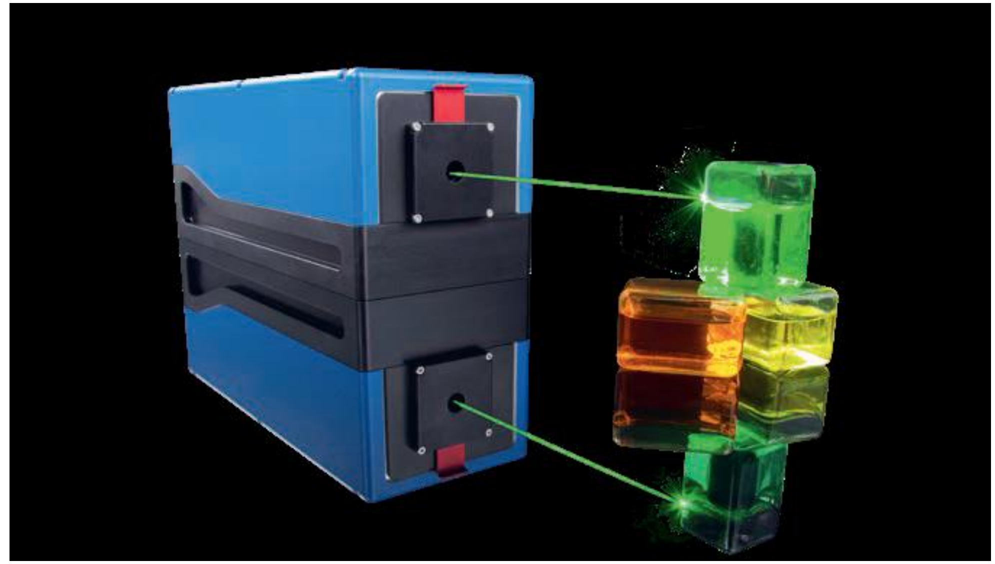
$$= \frac{1\ [\text{mol}]}{22400\ [\text{cm}^3]} \cdot \frac{9}{16} \cdot 1.672 \times 10^{-6} \times 2.997 \times 10^{10}\ [\text{cm/s}]$$

$$= 1.26\ [\text{mol/cm}^2/\text{s}]\ \ @\ 900\ [\text{K}]$$

With 2mmϕ nozzle → 0.068 mol/s =282kg/d (Ca total)
= 527g/d (^{48}Ca) = 192 kg/yr/unit
--- 6 units are necessary to produce 1 ton/yr.
Mean velocity of atoms is 868 m/s

Laser specification

(1) wavelength = 457nm
(2) average power = 400mW
(3) beam size = 2.5mm
(4) repetition rate = 190MHz
(5) pulse width = 2.63 ns
(6) duty factor = 50%



TOPTICA/DLC DL-pro-HP
with DLC TA-SHG pro option

Comparison of cost for ^{48}Ca enrichment

(Assuming total efficiency as 64%, so required laser power is not 190kW but 300kW)

	LIS (conventional)	**LISSE (new)**
# of units	3000	6
Lasers & Optics	18 billion JPY	42 million JPY
Electricity for laser	1.23 billion JPY	0.123 million JPY
Electricity for oven	0.345 billion JPY	0.345 billion JPY
Vacuum chamber	51 billion JPY	100 million JPY
Total cost	70.58 billion JPY	0.487 billion JPY
Unit cost	70580JPY/g	487JPY/g

Room to further cost reduction

~1/150

If we succeed in mass-production of enriched stable isotope, …

1) nuclear physicists do not need to prepare isotopes for targets and ion sources, $0\nu\beta\beta$ and n-nbar oscillation as well.
2) we can obtaine raw materials for nuclear medicine.
3) supply of ^{3}He for dilution refrigerator and fusion fuel in future
4) low-activable material by removing neutron-capturing isotopes
5) high-performance neutron shield by enriched ^{113}Cd
6) high-performance material. Ex. 99.998% ^{12}C enriched diamond for quantum computer
7) Passive tracer for studying material cycle on the Earth
8) Nuclear watermark (artificially modified isotopic abundances)

Summary

1) We propose a new method to efficiently separate stable isotopes with use of the Rabi oscillation induced by laser field.
Patent: application number 2023-149474

2) Test experiment to observe the Rabi oscillation will be performed in the first half of 2024FY using rubidium vapor and existing apparatuses for ^{3}He spin filter at Nagoya University.

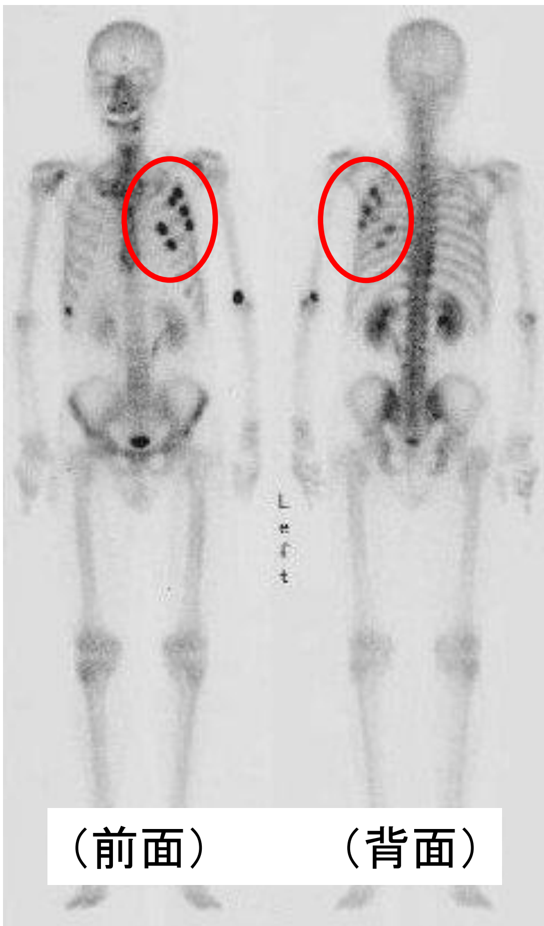
3) Technical key points;

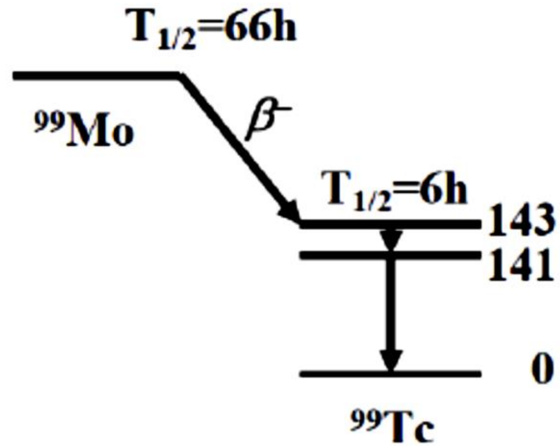
- Stability of the resonance of the cavity with $Q \sim 10^5$, needs a good feedback system for mirror distances.
- Quality and intensity of atomic beam is crucial.
  Ongoing R&D by CANDLES group (Rittirong, Ogawa) is important!

RI for medicine

$^{99}Tc^{m}$ for SPECT
(Single Photon Emission CT)

大腸癌の
骨転移例





^{99}Tc decays with $T_{1/2}$= 6hr emitting
141keV γ ⇒ measured by γ-camera

# of cases： 30 million/yr (world)
0.9 million/yr (Japan)
= 37000GBq/week imported

^{99}Mo is produced via thermal neutron capture.
^{nat}Mo target produces other radioactivities.
→ Using ^{98}Mo enriched target, only ^{99}Mo can be produced.

【核分裂法：mg オーダーの吸着】

^{99}Mo のみ吸着
⇒比放射能が高いことから，吸着させる Mo 量は微量で可能

【放射化法：g オーダーの吸着】

^{99}Mo だけでなく他の Mo 同位体も吸着
⇒比放射能が低いことから，吸着させる Mo 量は大量に必要

7days rxposure test at JRR-3 reactor, 1.85MBq/g was produced. To cover domestic demand (3.7×10^7MBq/week),

20tons of ^{nat}Mo or 5tons of ^{98}Mo are necessary every week.

Price of ^{nat}Mo; 212JPY/g

→ Production cost of ^{98}Mo should be less than ~700JPY/g.

For enrichment of Mo isotopes…

Atomic beam is not useful because of B.P. as high as 4639°C
MoF_6 can be vaporized at 34 °C.
Spectral data of MoF_6 is needed!