Nuclear Gamma-Ray Laser of Optical Range

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L.A. Rivlin

Nuclear gamma-ray laser: the evolution of the idea.

Quantum Electronics 37 (N8) 723-744 (2007)

The evolution of the foreign and native search for solving the problem of a nuclear gamma-ray laser (NGL), which has been attracting attention for almost half a century despite the absence at present of any convincing data about its experimental solution, is considered. It is shown that **the key conflict inherent in any conception of the NGL is the antagonism between the necessity to accumulate a sufficient amount of excited nuclei and the requirement to narrow down the emission gamma-ray line to its natural radiative width.** The critical analysis of different approaches for solving this coneict (Mossbauer scheme, deeply cooled ensembles of free nuclei with the hidden inversion, nuclear inversionless ampliecation, two-quantum gamma emission in counter-propagating photon beams, hypothetical amplifying medium of long-lived isomers in a Bose e Einstein condensate) shows that this search is important not only due to the expected result, which could stimulate the development of quantum nucleonics as a new branch in physics, but also is of interest due to a variety of physical disciplines and experimental approaches used in this search.

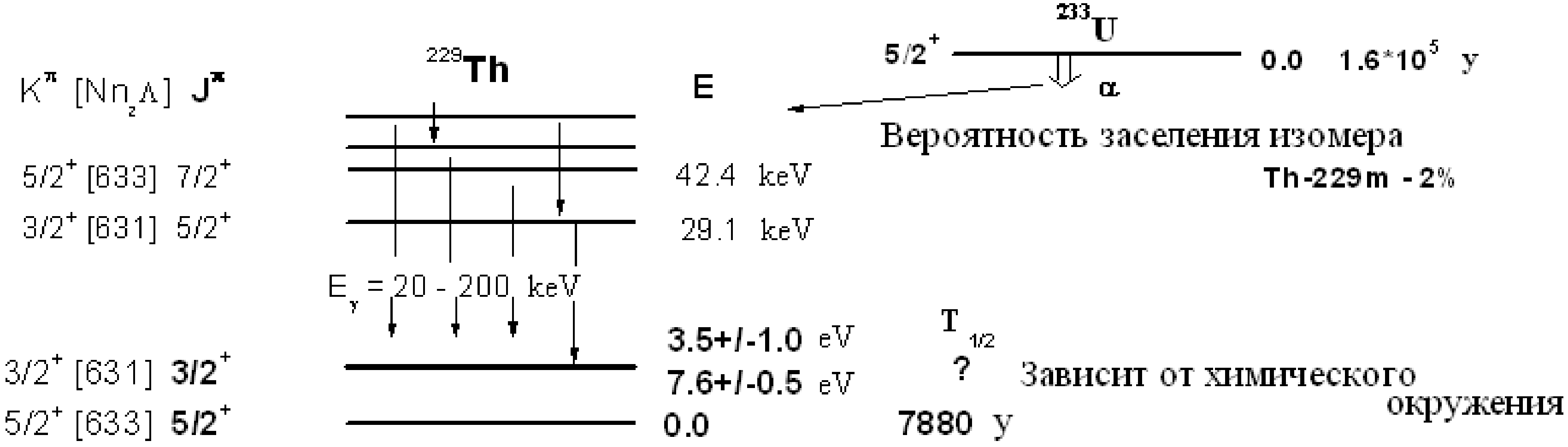
Ядерный γ-лазер оптического диапазона

Сформулированы условия при которых возможно усиление γ-излучения оптического (VUV) диапазона стимулированным излучением ансамбля изомерных ядер ^{229m}Th (3/2$^+$, 7.6 eV)

Усиление γ-излучения является результатом:

1) **возбуждения изомеров ^{229m}Th, размещенных в диэлектрике с большой шириной запрещенной зоны, лазерным излучением;**
2) **создания инверсной заселенности ядерных уровней в охлажденном образце вследствие взаимодействия ядер с внешним магнитным полем, или внутренним электрическим полем кристалла;**
3) **испускания и поглощения оптических фотонов ядром тория в кристалле без отдачи (эффект Мессбауэра в оптическом диапазоне);**
4) **спиновой релаксации (установления Больцмановского распределения заселенности) при взаимодействии ядра с электронами проводимости в металлическом покрытии.**

Энергия изомерного состояния



Вероятность заселения изомера
Th-229m - 2%

Зависит от химического окружения

Ключевые эксперименты

Анализ энергий и интенсивностей γ - переходов в ^{229}Th при α - распаде ^{233}U

1976	E_{is} < 100 eV	L.A.Kroger, C.W.Reich. *Nucl.Phys.* **A259** (1976)29
1989	E_{is} < 10 eV	INEL (USA)
1990	E_{is} < 5 eV	C.V.Reich, R.G.Helmer . *PRL* **64** (1990) 271
1994	E_{is} = 3.5+/-1.0 eV	R.G.Helmer, C.V.Reich. *PRC* **49** (1994) 1845
2007	E_{is} = 7.6+/-0.5 eV	B.R.Beck et al. *PRL* 98 (2007) 142501

Разрешение - 17 **эВ/канал**

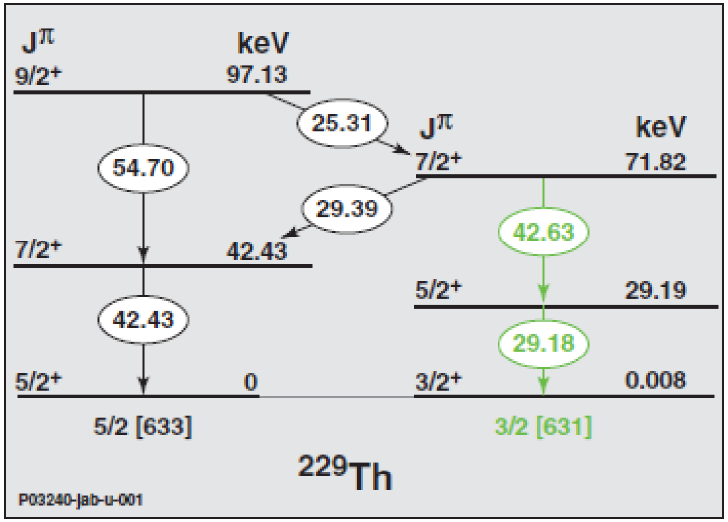
Ядерные реакции

1990 E_{is} < 6 keV D.G.Burke et al. *PRC* **42** (1990) R499

Реакция ^{230}Th(d,t)^{229}Th при E_d=17 MeV

McMaster University Tandem Accelerator. **Разрешение 6-7 кэВ**

Energy of the isomeric level

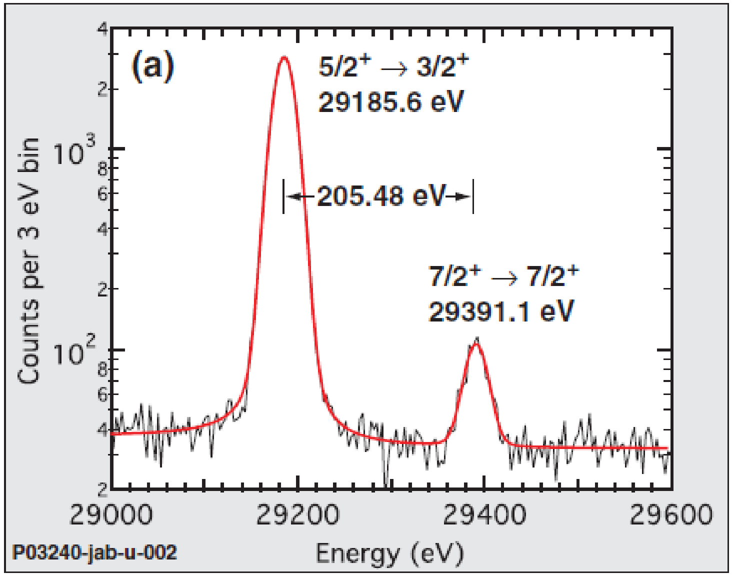


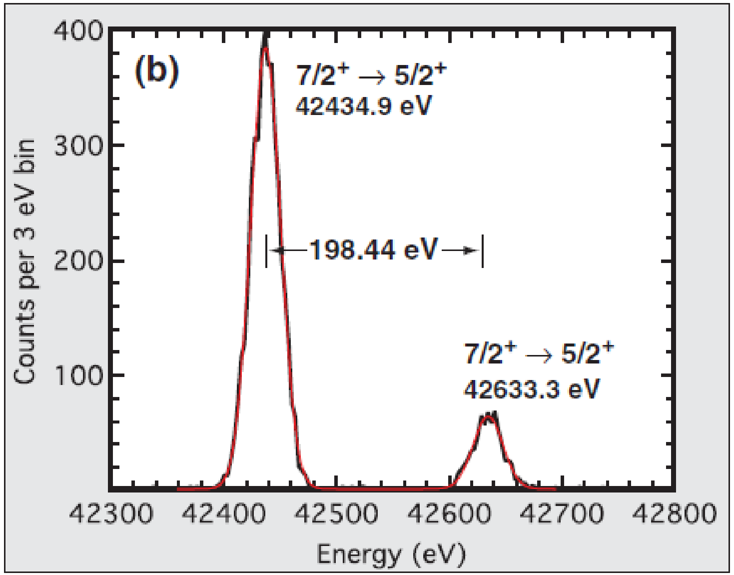
E_{is} = 7.6 +/- 0.5 eV

Phys.Rev.Lett. **98,** 142501 (2007)

B.R.Beck, J.A.Becker, P.Beiersdorfer et al.
Lawrence Livermore National Laboratory,
Los Alamos National Laboratory,
NASA Goddard Space Flight Center

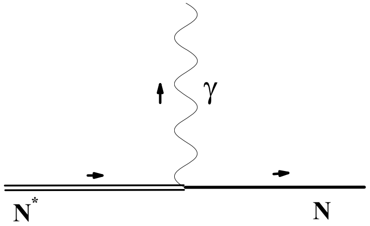
Источник: U-233 (105 μCi)
Детектор: NASA/electron beam ion trap x-ray microcalorimeter spectrometer
Разрешение: 26 eV (FWHM).





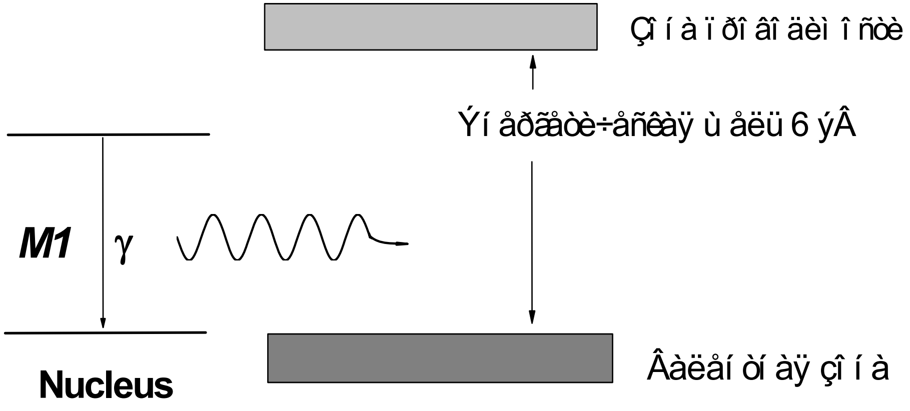
Канал распада в диэлектрике

"Ядерный свет"



Е.В.Ткаля. *Вероятность спонтанного излучения для М1 перехода в диэлектрической среде: распад ^{229m}Th(3/2+, 3.5±1.0 эВ)* . Письма в ЖЭТФ **71** (2000) 449.

E.V.Tkalya et al. *Decay of the low-energy nuclear isomer ^{229m}Th(3/2+, 3.5+/-1.0 eV) in solids (dielectrics and metals): A new scheme of experimental research*. Phys.Rev.C **61** (2000) 064308.



Äâóîêèñü òîðèÿ - ^{229}ThO$_2$

Время жизни уровня при E_{is} = 3.5+/-1.0 eV:
10 мин - 1 час

А.М.Дыхне, Е.В.Ткаля. *Матричный элемент перехода аномально низкой энергии 3.5±0.5 эВ в ядре ^{229}Th и время жизни изомера*. Письма в ЖЭТФ **67** (1998) 233.

$B(M1)_{Wu} = 4.8 \times 10^{-2}$ с учетом кориолисова взаимодействия

Зависимость вероятности спонтанного М1 γ распада от показателя преломления среды "n":

$$W_{medium} = n^3 W_{vacuum}$$

Structure and parameters of $LiCaAlF_6$

S. Kuze et al. J. Solid State Chem. **177** (2004) 3505

S. Kuck et al. Laser Phys. **11** (2001) 116

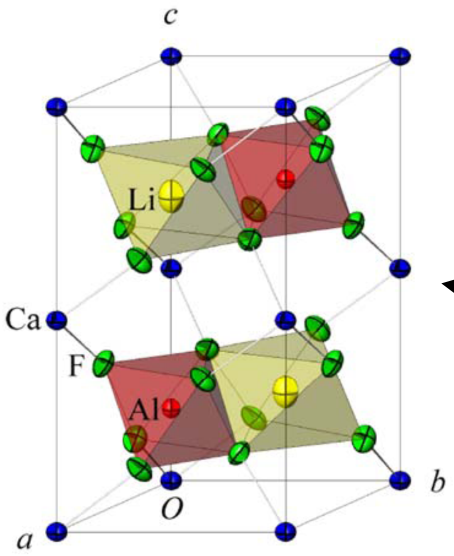
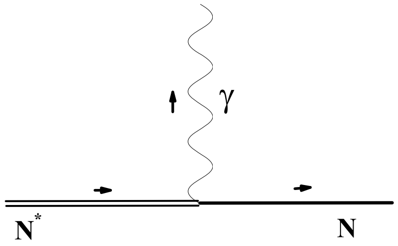


Fig. 1. The structure of LiCAF at 300 K with Ca (blue), Al (red), Li (yellow) and F (green) atoms.

Symmetry: Trigonal
Band gap: 110 nm (**11.1 eV**)
Melting temperature: 825°C

$^{229}Th{:}LiCaAlF_6$

"The Nuclear Light" $\omega_\gamma = 7.6$ eV



$T_{1/2} \approx 25$ min
if the refractive index $\varepsilon^{1/2} = 1$

The amplification coefficient

$$\boxed{\chi = \frac{\lambda_{is}^2}{2\pi}\frac{\Gamma_{rad}}{\Delta\omega_{tot}}\frac{1}{1+\alpha}\left(n_{is} - \frac{n_{gr}}{g}\right) - \kappa \qquad \text{cm}^{-1}}$$

$$E_{is} = 7.6 \pm 0.5 \quad \text{eV}$$

$$g = \frac{2J_{gr}+1}{2J_{is}+1} = 1.5$$

$$\lambda_{is} = 163 \pm 11 \quad \text{nm}$$

$$\Gamma_{rad}(M1; is \to gr) \approx 3\times 10^{-19} \ \text{eV}$$

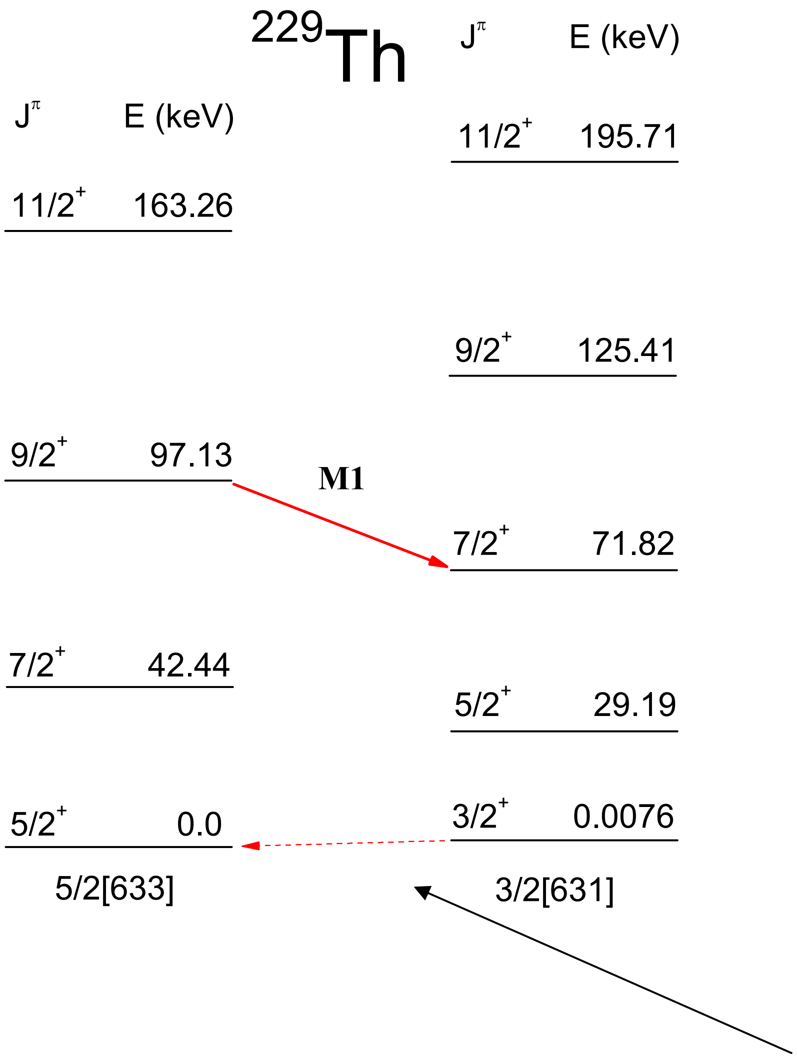
$$T_{1/2}^{is} \approx 25 \quad \text{min}$$

W.G. Rellergert et al. Phys.Rev.Lett. **104**, 200802 (2010).

The sample ^{232}Th:LiCAF , n (^{232}Th) = 10^{18} cm^{-3}

^{229m}Th:LiCaAlF$_6$

$$\Delta\omega_{tot} \le 7\times 10^{-13}\,\text{eV} \ \ (=1\ \text{kHz}) \qquad \alpha = 0 \qquad \kappa \approx 0.01\ \text{cm}^{-1}$$



The *M*1 transition
9/2+(97.13 keV)→7/2+(71.82 keV):
$B(M1)_{\mathrm{W.u.}} = 0.038,\ 0.024,$ and 0.014

The Coriolis interaction between rotational bands enhances the transition probability by a factor of 1.2–1.3

The ‘‘enhanced’’ average value for the transition
3/2+(7.6 eV)→5/2+(0.0):
$B(M1)_{\mathrm{W.u.}} = 0.032$

The value of the radiative width: $\Gamma_{\mathrm{rad}} = 3\times10^{-19}$ eV

^{229}Th:LiCaAlF$_6$

	The Abundance Ratio of the Isotopes, %		Spin	The ground state	
				μ/μ_N	Q, eb
Li :	^{6}Li	7.5	1+	+0.822	-0.818×10^{-3}
	^{7}Li	92.5	3/2-	+3.2564	-0.0406
Ca:	^{40}Ca	96.94	0+		
	^{42}Ca	0.647	0+		
	^{43}Ca	0.135	7/2-	-1.31726	-0.043
	^{44}Ca	2.09	0+		
	^{46}Ca	0.004	0+		
	^{48}Ca	0.187	0+		
Al:	^{27}Al	100	5/2+	+3.64	+0.1402
F:	^{19}F	100	1/2+	+2.6289	
Th:	^{229}Th		5/2+	+0.46	+4.3
	^{232}Th	100	0+		

Excitation of ^{229m}Th(7.6 eV) by laser radiation

$$dn_{is}/dt = \sigma\varphi n_{gr} - \Lambda_{is} n_{is} - g\sigma\varphi n_{is}$$

$$dn_{gr}/dt = -\sigma\varphi n_{gr} + \Lambda_{is} n_{is} + g\sigma\varphi n_{is}$$

Initial Conditions

$$n_{is}(0) = 0$$

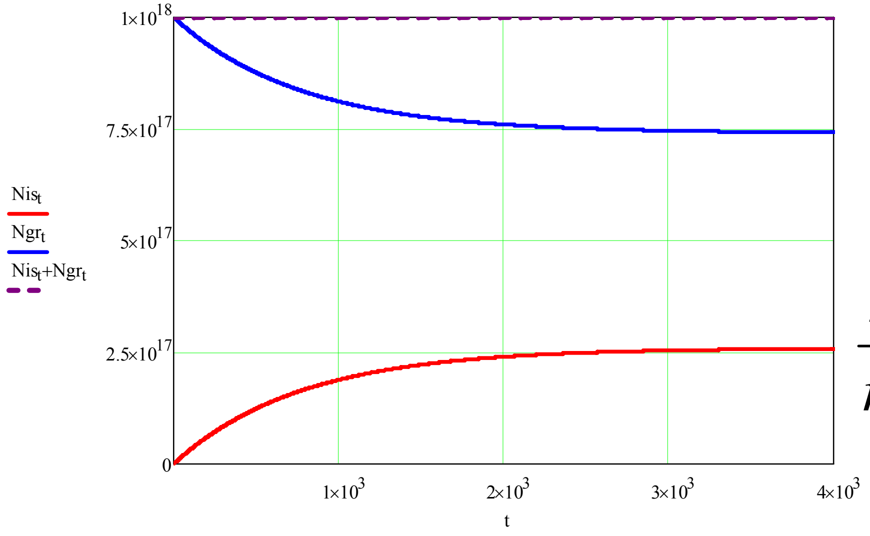
$$n_{gr}(0) = 10^{18} \qquad \text{cm}^{-3}$$

$$\sigma = \frac{\lambda_{is}^2}{2\pi}\frac{\Gamma_{rad}}{\Delta\omega_L}\frac{1}{g} \approx 10^{-24} \quad \text{cm}^2$$

$$\Delta\omega_L/\omega_L = 10^{-6}$$

$$\Lambda_{is} = \Gamma_{rad} = \ln 2 / T_{1/2}^{is}$$

$$\varphi \approx 10^{20} \quad \text{cm}^{-2}\ \text{s}^{-1}$$



$$\frac{n_{is}}{n_{gr}} \approx \frac{1}{3}$$

$$t \approx (2-3)T_{1/2}^{is}$$

VUV Lasers

The energy of the isomeric level is known roughly, and we can not tell now, what VUV laser will be used for the pumping of the ^{229m}th isomers.

1. We can use one of the available lasers (see in *Springer handbook of atomic, molecular, and optical physics*. G.W.F. Drake (Ed.), Springer, 2nd ed., 2006).

   Commercially available lasers have the power P = 1-3 W.

   Molecular CO and H_2 lasers span region around 164 nm.
2. It will be necessary to develop a special laser with the corresponding wavelength.
3. We can use a free electron laser (such lasers have a good tunability).

For irradiation of the sample we need density of the photon flux 10^{20} cm^{-2} s^{-1}. Such flux density can be reached relatively easily by focusing of the radiation of middle power laser.

The Mossbauer effect in the optical range

The energy lost E_R due to the recoil is negligibly small:

$$E_R = \omega^2 / 2M \quad = 1.4\times10^{-10}\ \mathrm{eV}$$

(M is the Th-229 nucleus mass, $\omega = 7.6$ eV)

The Debye-Waller factor $f \approx \exp(-3E_R / 2\theta_D) \quad = 1$

because $E_R/ \theta_D << 1$ (θ_D is the Debye temperature)

Emission of the γ–ray photons by the ^{229m}Th isomers and the resonant absorption of these photons by the ^{229}Th nuclei in a solid should occur without recoil.

Splitting in external magnetic field

Populations of the Zeeman sublevels are described by

$$\exp(-\mu_{gr(is)}H/T)$$

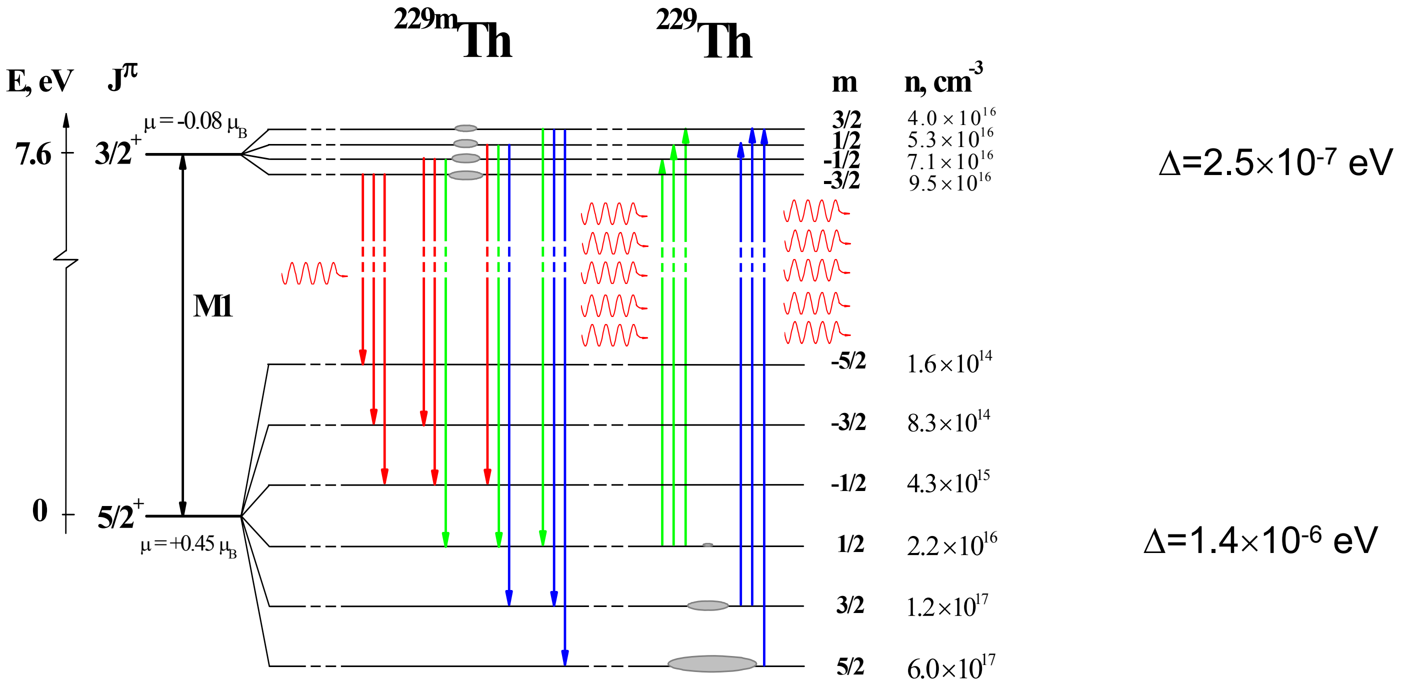
The magnetic moment of the ground state $\mu_{gr} = 0.45\mu_N$
The magnetic moment of the isomeric state
(theoretical estimation) $\mu_{is} = -0.08\mu_N$

μ_N is the nuclear magneton

The population of the ground state sublevels falls down much faster, than the populationof the isomeric sublevels because

$$|\mu_{gr}/\mu_{is}| \approx 6$$

Splitting in magnetic field $H = 100$ T at $T = 0.01$ K



Population of the sublevels of the ground state and the isomeric state corresponds to the following case:

Laser: $P = 30$ mW
Density of Th-229: $n_{gr}(0) = 10^{18}$ cm^{-3}

The amplification coefficient for "**red**" transitions

$$\chi \approx 3 \text{ cm}^{-1}$$

Structure and parameters of $LiCaAlF_6$

J.B. Amaral et al. J. Phys.: Condens. Matter **15** (2003) 2523

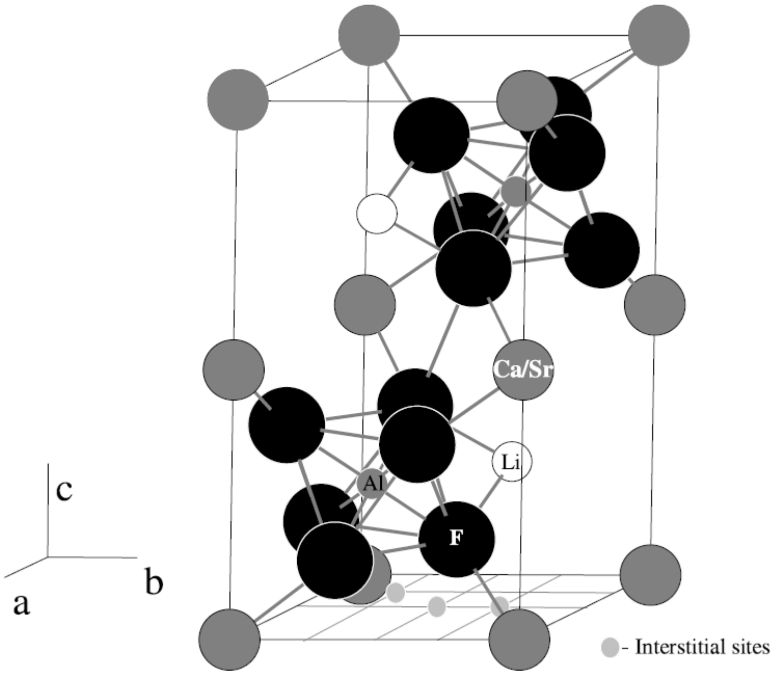


Figure 1. Unit cell representation of the $LiCaAlF_6$/$LiCaAlF_6$. The grey circles indicate the positions of the three interstitial sites.

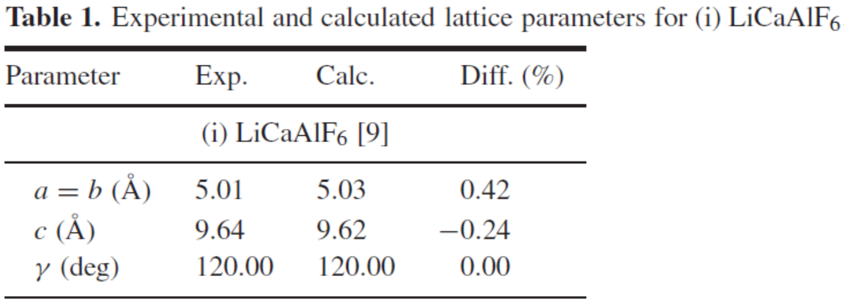


Table 1. Experimental and calculated lattice parameters for (i) $LiCaAlF_6$

Parameter	Exp.	Calc.	Diff. (%)
	(i) $LiCaAlF_6$ [9]		
$a = b$ (Å)	5.01	5.03	0.42
c (Å)	9.64	9.62	−0.24
γ (deg)	120.00	120.00	0.00

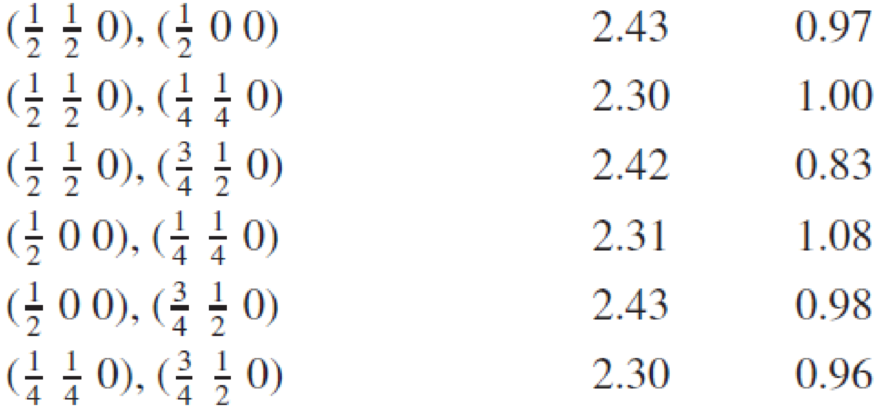
Structure of $^{229}Th:LiCaAlF_6$

R.A. Jackson et al. J. Phys.: Condens. Matter **21** (2009) 325403

Computer modelling of thorium doping in $LiCaAlF_6$

Site	**Reaction**
Ca^{2+}	$ThF_4 + Ca_{Ca} \rightarrow Th^{\cdot\cdot}_{Ca} + 2F_I^- + CaF_2$

Solution energies (eV) for Th^{4+} doping in LiCAF



Interstitial positions	Unbound	Bound
$(\frac{1}{2}\ \frac{1}{2}\ 0), (\frac{1}{2}\ 0\ 0)$	2.43	0.97
$(\frac{1}{2}\ \frac{1}{2}\ 0), (\frac{1}{4}\ \frac{1}{4}\ 0)$	2.30	1.00
$(\frac{1}{2}\ \frac{1}{2}\ 0), (\frac{3}{4}\ \frac{1}{2}\ 0)$	2.42	0.83
$(\frac{1}{2}\ 0\ 0), (\frac{1}{4}\ \frac{1}{4}\ 0)$	2.31	1.08
$(\frac{1}{2}\ 0\ 0), (\frac{3}{4}\ \frac{1}{2}\ 0)$	2.43	0.98
$(\frac{1}{4}\ \frac{1}{4}\ 0), (\frac{3}{4}\ \frac{1}{2}\ 0)$	2.30	0.96

Quadrupole splitting.

Spectroscopic quadrupole moments

The **ground state** spectroscopic quadrupole moment
Q_{gr} = 3.149±0.032 eb **(4.3±0.9 eb - from the optical determinations)**.

We make the standard assumption that the intrinsic quadrupole moment **Q_2 = 8.816 eb** remains the same for the rotational bands $K = 5/2$ and $K = 3/2$ in Th-229.

$$Q_{is} = \frac{3K_{is}^2 - J_{is}(J_{is}+1)}{(J_{is}+1)(2J_{is}+3)} Q_{20}$$

Then the **isomeric state** spectroscopic quadrupole moment
Q_{is} = 1.8 eb (**2.4 eb**).

Quadrupole splitting.

The electric field gradient (EFG)

“Wien2k”: EFG at the Ca^{2+} ion site in LiCAF is

$$\varphi_{zz} = -\,1.2\times 10^{17}\ \mathrm{V/cm^2}$$

In the ^{229}Th:$LiCaAlF_6$ crystal the leading contribution to EFG at the Th^{4+} ion site comes from F^- ions, which compensate the extra charge 2+.

These ions are located in interstitial sites in the vicinity of Th^{4+}. An estimation gives

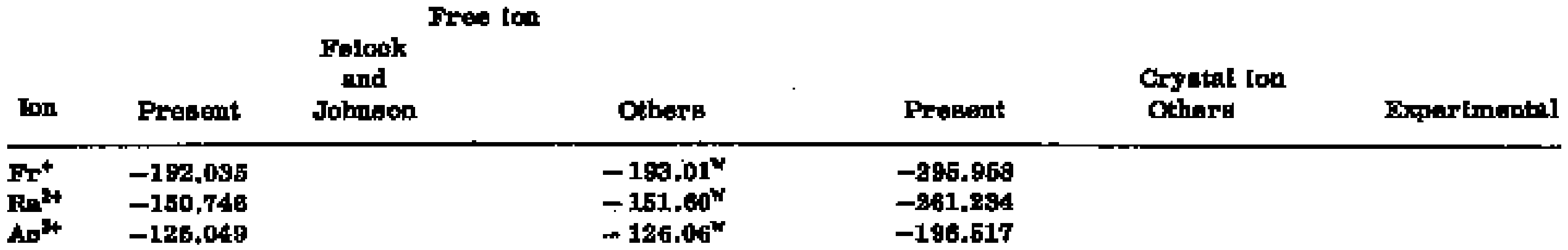
$$\varphi_{zz} \approx -\,10^{18}\ \mathrm{V/cm^2}$$

at the Th^{4+} site.

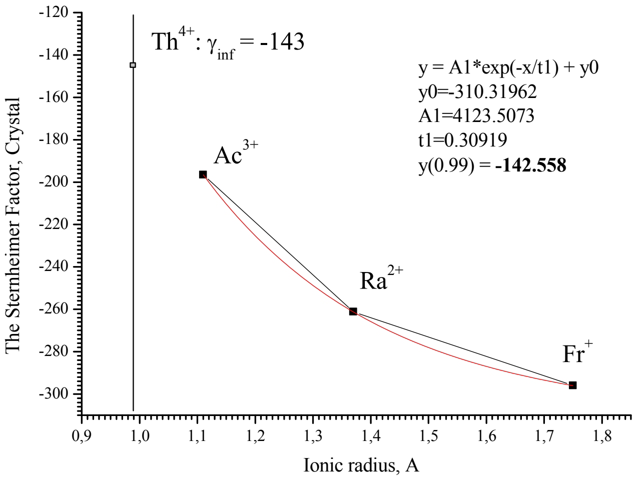
Quadrupole splitting.

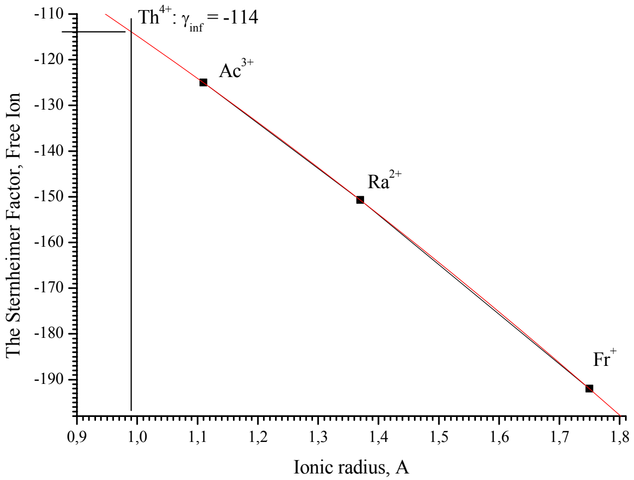
The Sternheimer antishielding factor

K.D. Sen and P.T. Narasimhan. *Quadrupole antishielding factors and polarizabilities in ionic crystals*. Phys.Rev.B (Solid State) **15** (1977) 95.



Ion	Free ion			Crystal ion		
	Present	Feiock and Johnson	Others	Present	Others	Experimental
Fr^+	−192,035		-193.01^w	−295.958		
Ra^{2+}	−150,746		-151.60^w	−261.234		
Ac^{3+}	−126,049		-126.06^w	−196.517		

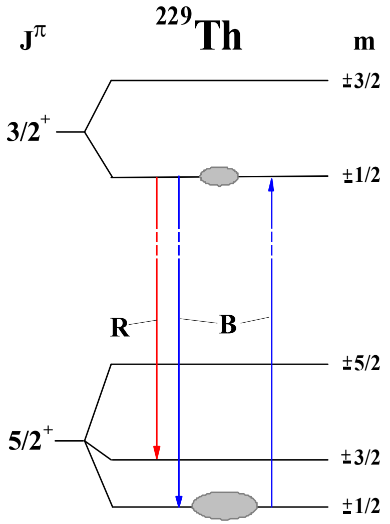




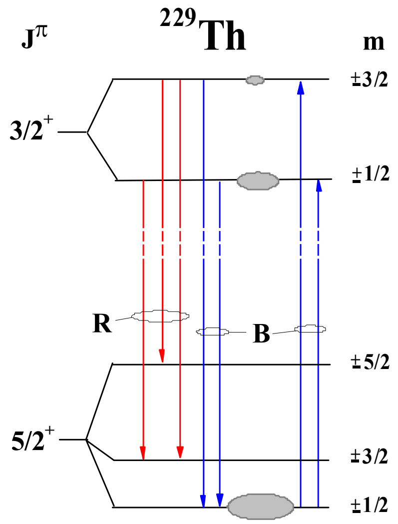
Th^{4+}

$$\gamma_\infty \approx -100$$

Quadrupole splitting



(a)



(b)

The sublevels energies are given by

$$E_m = eQ_{gr(is)}(1-\gamma_\infty)\varphi_{zz}\frac{3m^2 - J_{gr(is)}(J_{gr(is)}+1)}{4J_{gr(is)}(2J_{gr(is)}-1)}$$

|is> $\Delta = 3.6\times10^{-5}$ eV

|gr> $\Delta = 4\times10^{-5}$ eV

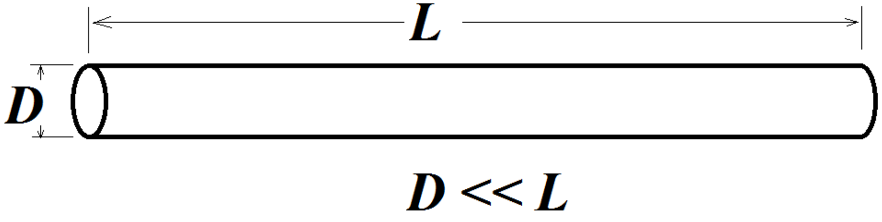
$\Delta = 2\times10^{-5}$ eV

The amplification coefficient
(a) $\chi \approx 2$ cm^{-1} (b) $\chi \approx 3$ cm^{-1}

At large values of EFG the effective inverse population and the amplification condition will hold up to the temperature $T = 0.1$ K.

Duration of the γ-ray laser emission, τ

$\tau \approx 10^2$ s if $D = 10^{-2}$ cm, $L = 5$ cm, $\chi = 3$ cm^{-1}



$$\tau \approx T^{is}_{1/2}(L/D)^2 \exp(-\chi L)$$

$$\tau << T^{is}_{1/2}, \quad L << \chi^{-1}\ln(N_{is}/2)$$

The Fresnel number $F = \pi D^2/4L\lambda_{is}$

$$N_{is} = n_{is}\frac{\pi D^2}{4}L \approx 4\times 10^{13}$$

$F = 10$ if $L = 0.5$ cm, $F = 1$ if $L = 5$ cm

Число Френеля характеризует дифракционные потери резонатора

The emission will be a sequence of pulses with the repetition frequency

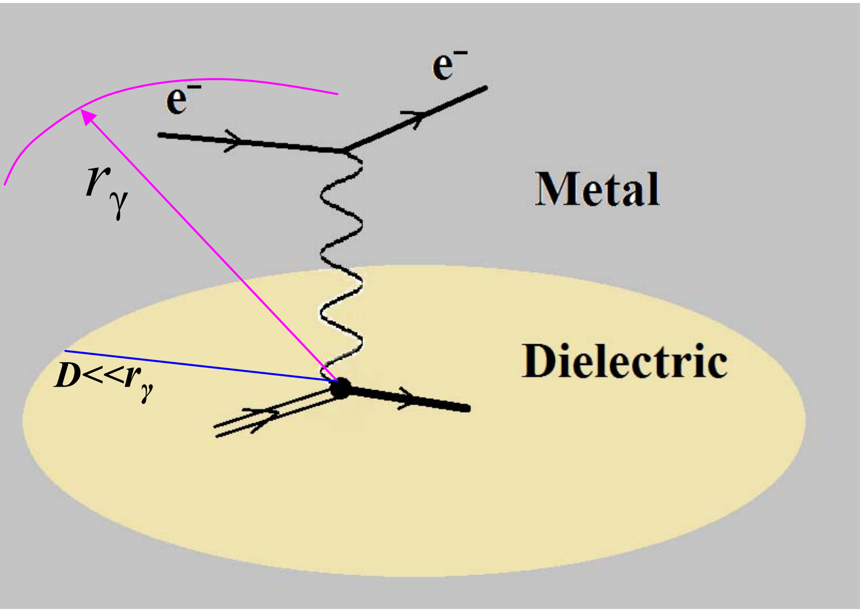
$f_{rep} = Q_{is}(D/L)^2 \approx 10^5 - 10^4$ s^{-1} where $Q_{is} = \dfrac{\ln 2}{T^{is}_{1/2}}N_{is} \approx 2\times 10^{10}$ s^{-1}

The mean power of the γ-ray laser will be $P \approx 10^{-7}$ W.

The gain: $\exp(\chi L) \approx 3\times 10^6$

Nuclear spin relaxation process.

Inelastic scattering of the conduction electrons on nucleus or “internal” conversion on the conduction electrons



Energy of the Zeeman (or quadrupole) splitting of nuclear sublevels

$$\Delta E = 10^{-6} - 10^{-7} \quad \text{eV}$$

Energy of the conduction electrons

$$E_e = E_F \approx 5.5 \qquad \text{eV}$$

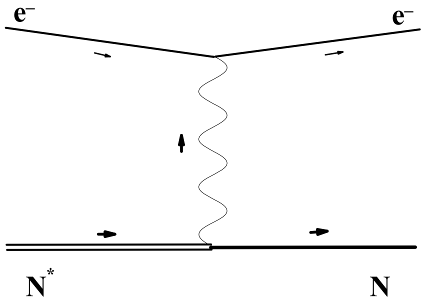
$$p_i = \sqrt{2m_e E_F} \qquad p_f = \sqrt{2m_e (E_F + \Delta E)}$$

The virtual photon

$$\omega_\gamma = \Delta E \qquad \vec{q}_\gamma = \vec{p}_f - \vec{p}_i \to q_\gamma^{\min} = \Delta E \sqrt{m_e / 2E_F} \qquad ƛ_\gamma = \frac{1}{q_\gamma^{\min}} \approx 0.1 - 1 \ \text{cm}$$

$$m_\gamma^* = \sqrt{q_\gamma^2 - \omega_\gamma^2} \approx q_\gamma^{\min} \qquad \Delta t \approx \hbar / m_\gamma^* \qquad \boxed{r_\gamma = c\Delta t \approx 1/m_\gamma^* \approx 0.1 - 1 \ \text{cm} \, !}$$

Распад изомерного состояния в металле



Е.В.Ткаля. *Безрадиационный распад низколежащего ядерного изомера ^{229}Th(3.5 эВ) в металле.* Письма в ЖЭТФ **70** (1999) 367.

Металл: конверсия на электронах проводимости (неупругое рассеяние электронов проводимости на ядрах). Энергетический порог у реакции отсутствует.

Время жизни изомера в "стандартном" металле < 1 с

Сечение неупругого рассеяния

Взаимодействие

$$H_{\text{int}} = e^2 \iint d^3 r\, d^3 R\, \overline{\psi}_f(\vec{r})\gamma^{\mu}\psi_i(\vec{r}) g_{\mu\nu} \frac{e^{i\omega|\vec{r}-\vec{R}|}}{|\vec{r}-\vec{R}|} \Psi_f^{+}(\vec{R}) \hat{J}_N^{\nu} \Psi_i(\vec{R})$$

ВФ электрона – плоские волны

Сечение

$$\sigma_{M1} = \frac{32\pi^2}{9} e^2 \ln\frac{4E_e}{\Delta E} B(M1)$$

при

$$\Delta E << E_e << m_e$$

Nuclear spin relaxation process

The "spin-lattice" relaxation time *T_1*

$$\frac{1}{T_1} \approx n_e \frac{\Delta E}{E_F} \sigma_e \xi_D \upsilon_F$$

The conduction electron density (**Au**, **Cu**, **Ag**) $n_e \approx (6 \div 8) \times 10^{22}$ cm^{-3}

$$\sigma_e(M1,\ \mu = 0.45\mu_N,\ \Delta E = 10^{-6} \div 10^{-7}\ eV) \approx 10^{-30} \quad \mathrm{cm}^{-2}$$

$$\upsilon_F = \sqrt{2E_F / m_e} \approx 4.6 \times 10^{-3} \qquad \xi_D \equiv \sigma_e(D)/\sigma_e$$

$$\xi_D(D = 0.01\,cm) \geq 0.1 \qquad \boxed{T_1 \leq 50 \ \ \mathrm{d}}$$

E.Klein, *Relaxation Phenomena*. In: *Low-Temperature Nuclear Orientation*. Eds. N.J.Stone and H. Postma, (North-Holland, Amsterdam, 1986) p.579.

…In insulators without electronic moments… (i.e. in pure crystals)… at millikelvin temperatures… *T_1* would exceed the age of the Universe…

Что делать?

1. Определить энергию изомерного уровня с точностью $\sim 10^{-6}$ эВ

a) На пучке СИ, возбуждая изомер через промежуточный уровень 5/2+(29.19 keV) в диэлектрической матрице;

b) На оптическом синхротроне прямым возбуждением изомера.

2. Изготовить образец ^{229}Th:LiCAF.

2. Измерить время спиновой релаксации T_1 в ^{229}Th:LiCAF.

3. Измерить величину квадрупольного расщепления в ^{229}Th:LiCAF.

4. **Исследовать возможность влияния металлического покрытия на спиновую релаксацию в диэлектриках при криогенных температурах.**

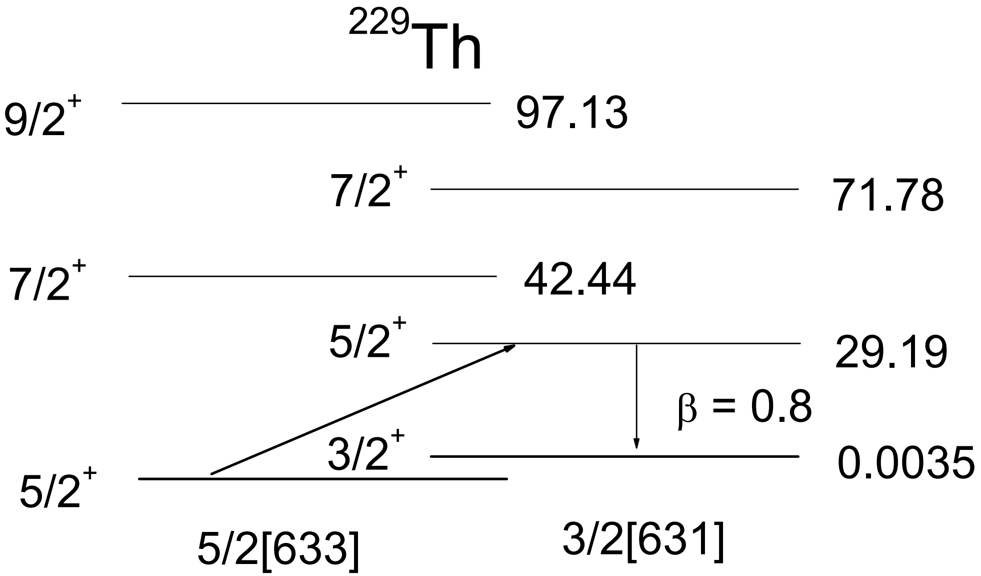
5. Подобрать лазер и осуществить «накачку» изомера.

И т.д.

Определение энергии изомерного уровня

Самый правильный эксперимент - возбуждение через промежуточный уровень синхротронным излучением

E.V.Tkalya et al. *Decay of the low-energy nuclear isomer 229mTh(3/2+, 3.5+/-1.0 eV) in solids (dielectrics and metals): A new scheme of experimental research*. Phys.Rev.C **61** (2000) 064308.

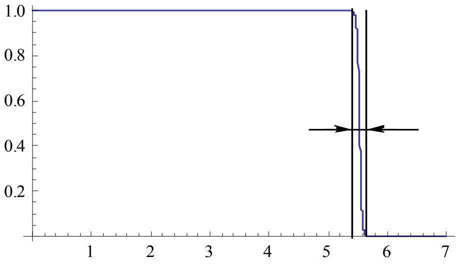


Advanced Photon Source at ANL
Ток I = 100-300 mA,
Энергия электронов E = 7 GeV,
Критическая энергия ω_C = 32.6 keV

Скорость возбуждения:
10^6 ядер/с в мишени из 1 мг ^{229}Th

Nuclear spin relaxation process

The Fermi-Dirac distribution for electrons



$\Delta E \approx T$

$$f_e(E) = \frac{1}{\exp\left(\frac{E - E_F}{T}\right) + 1}$$

Nuclear Gyroscope

^{93}Nb: $I^{\pi}=9/2^{+}$, $\mu = 6.17\ \mu_N$

$H = 10$ T

$T = 0.02$ K

Polarization

$$E_i = \mu H m_i \qquad n_i = e^{-\frac{E_i}{kT}} / \sum_i n_i \qquad f_1 = \frac{1}{I}\sum_i m_i n_i = 0.894$$

^{93}Nb$_2$O$_5$ - dielectric, $\Delta \approx 3.9$ eV

$\sigma_{M1} \approx 10^{-28}$ cm^2 $\quad T_1 \sim 1$ h