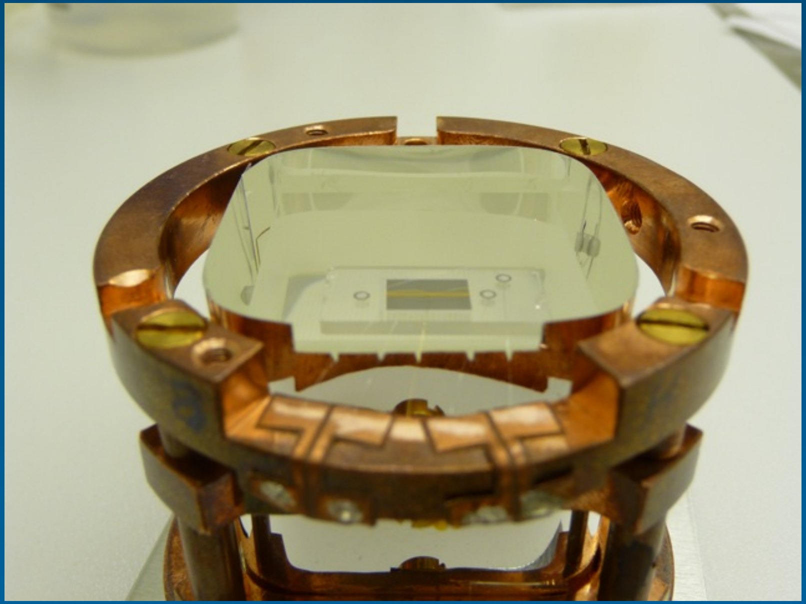
Scintillating $CaWO_4$ Crystals for CRESST-II and EURECA



M. v. Sivers, Kiev 19.09.2013

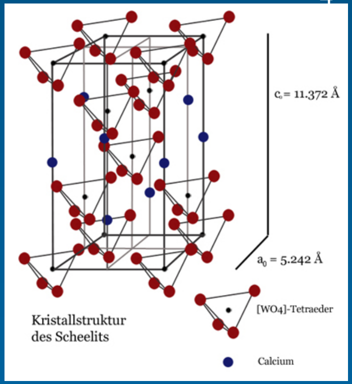
$CaWO_4$ Crystals for Rare Event Searches

- Current target material in CRESST-II (since July 2013 ≈5.5kg)
- Planned as target material for EURECA (Phase I≈75kg, Phase II≈500kg)
- Suppliers: GPI RAS (Russia), SRC “CARAT” (Ukraine), …
- **In house production at TUM since 2011**
- Multi-material target
  - W (A=184): Good sensitivity for coherent scattering ($\sigma_{coh} \sim A^2$)
  - Ca, O: Sensitivity to light WIMPs
  - ^{183}W (14%): some sensitivity to spin-dependent scattering
  - ^{48}Ca (0.2%): candidate for 0ν2β

Crystal Properties

Property	Value
Density	6.1g/cm^3
Melting point	1600°C
Hardness	4.5-5 Mohs
Cleavage planes	distinct on {101}, indistinct on {001}
Space group	$I4_1/a$
Cell parameters	a=5.242Å, c=11.372Å

Unit cell of $CaWO_4$

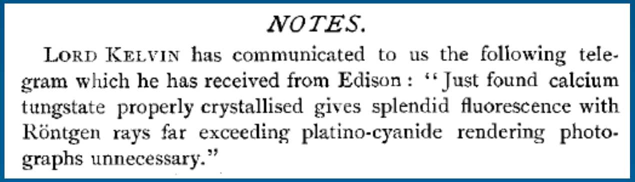


Naturally occurring scheelite crystal (China)

Optical and Scintillation Properties

Property	Value
Refractive index	1.95
Birefringence	δ≈0.017
Luminescence center	WO_4^{2-}
Emission maximum	420nm
Light Yield (at RT)	16,000-20,000ph/MeV[1,2] (≈40% of NaI(Tl))
Decay time (at RT)	9µs

Nature 53, 470 (1896)

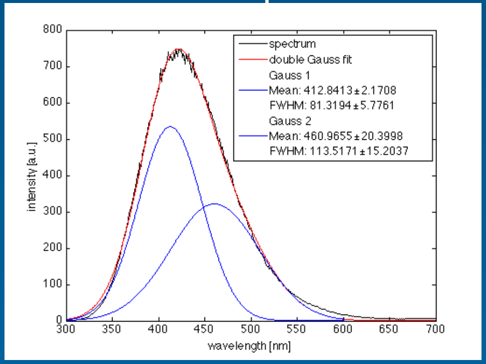


NOTES.

LORD KELVIN has communicated to us the following telegram which he has received from Edison: "Just found calcium tungstate properly crystallised gives splendid fluorescence with Röntgen rays far exceeding platino-cyanide rendering photographs unnecessary."

$CaWO_4$ under UV excitation

Emission spectrum

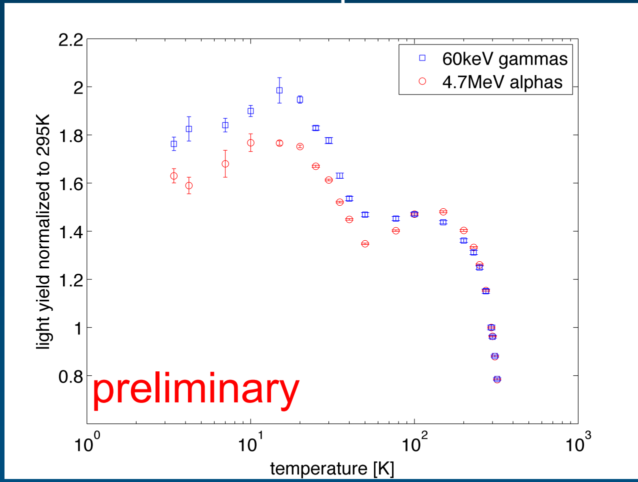


[1]M. Moszynski et al., NIM A 553, 578-591
[2]M. v. Sivers et al., Opt. Mat. 34, 1843–1848

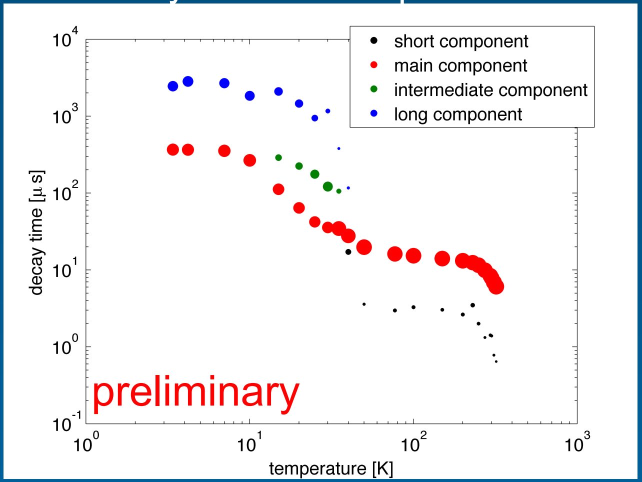
Scintillation Properties at Low Temperatures

- ≈70% increase of LY at low temperatures
- Increase of main decay time to ≈350µs, appearance of long decay component (≈2ms)
- PSD between alpha and gamma particles possible
- α/γ quenching factor ≈0.22

LY vs. temperature

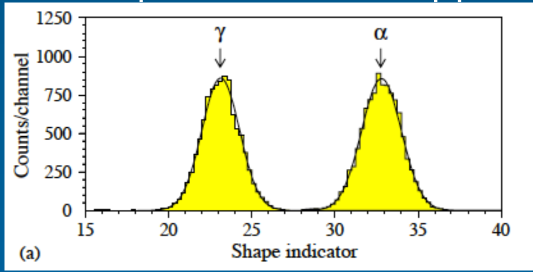


Decay time vs. temperature



Measurements in collaboration with P. Di Stefano (Queen's University)

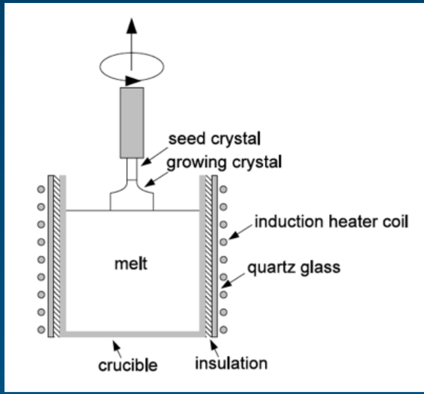
Pulse shape indicator for α/γ particles

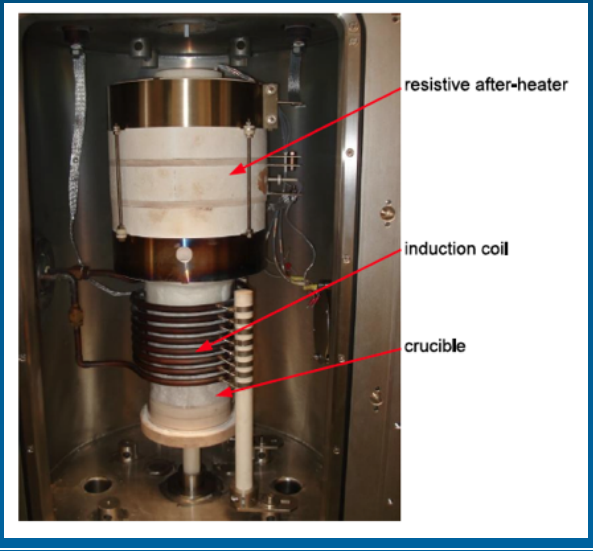


Y. Zdesenko et al., NIM A 538, 657-667

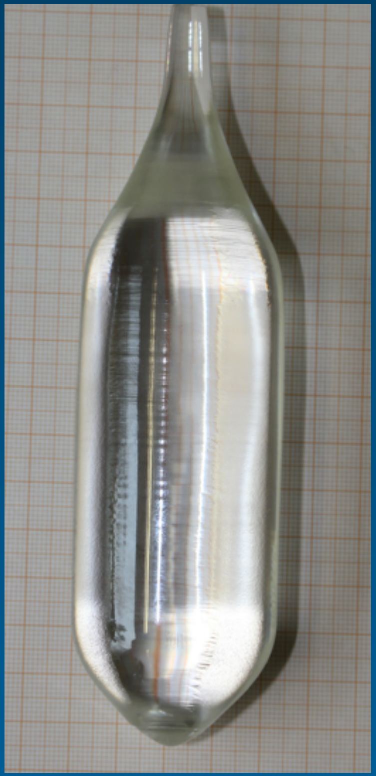
Czochralski Growth at TUM

- Cyberstar Oxypuller 20-04 Czochralski furnace
  - 80mm/120mm diameter Rh crucibles
  - Continuous weighing of crystal during growth → automatic adjustment of heating power by PI controller
  - After-heater on top of crucible → lower risk of cleavage due to thermal stress during cooling
  - Growth under flow of 99% Ar, 1% O_2 → reduce oxygen deficiency of crystals





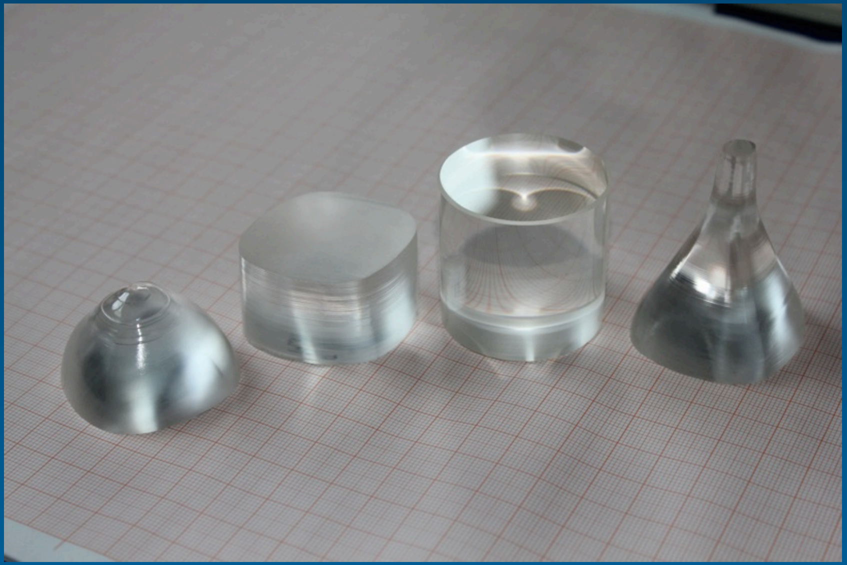
Grown Ingots



Raw ingot "Bernhard I"

- m=890g
- d=44mm, h=65mm (cylindrical part)

Sawing and polishing



Cylindrical detector crystal

- m=300g
- d=40mm, h=40mm

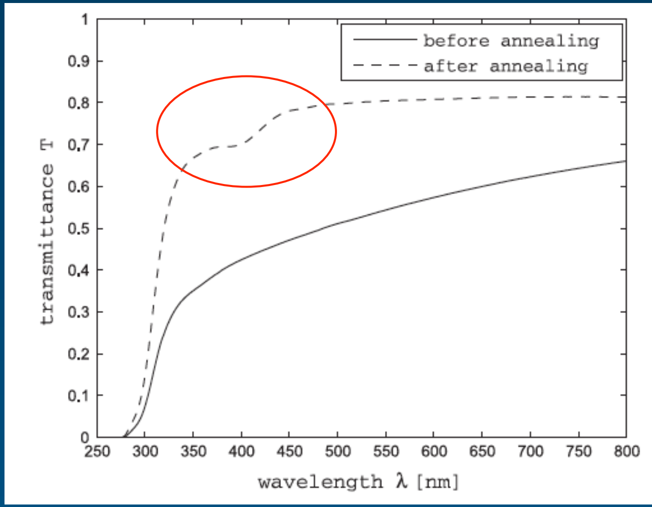
Annealing Process

- After-growth annealing under O_2-flow at 1450°C, 72h
  - Reduction of internal stresses
  - Filling of oxygen vacancies

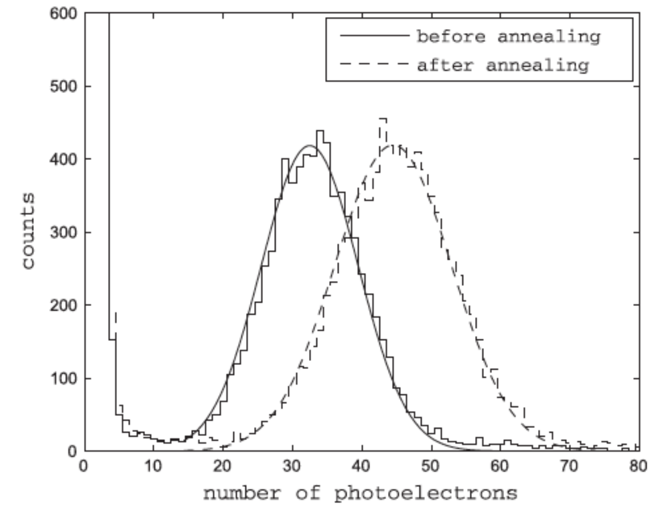
$$V_O^{\bullet\bullet} + \frac{1}{2}O_2 \rightarrow O_O^{\times} + 2h^{\bullet}$$

  - ➢ Decrease of absorption coefficient by factor ≈6
  - ➢ Increase of light yield by ≈40%
  - ➢ Absorption band @400nm possibly due to O^- centers

Transmission measurement

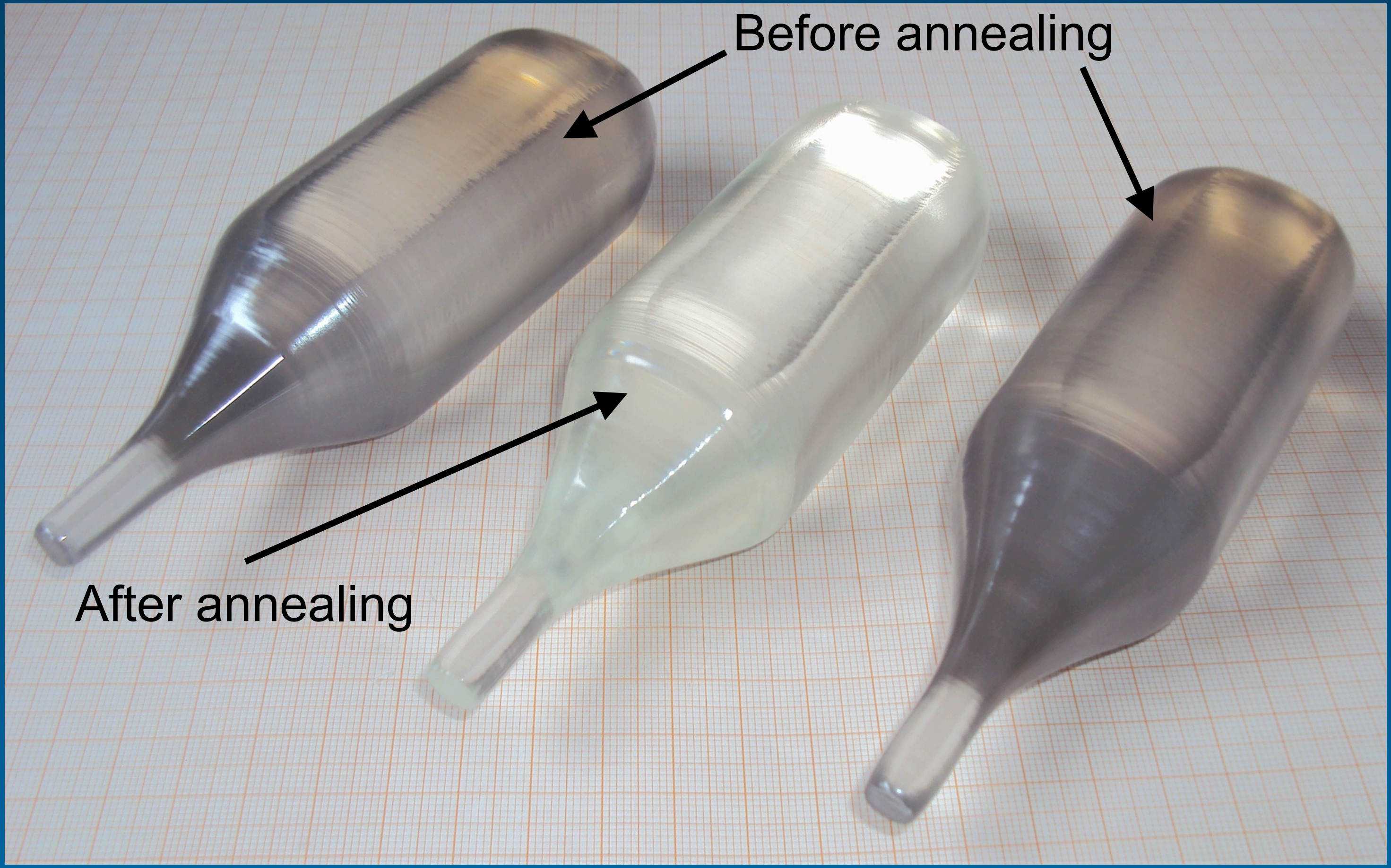


^{241}Am spectrum (59.5keV)



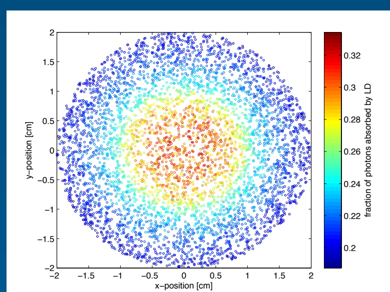
M. v. Sivers et al., Opt. Mat. 34, 1843–1848

Influence of Annealing

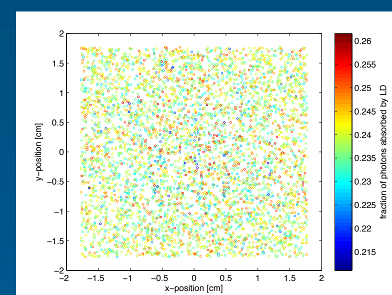


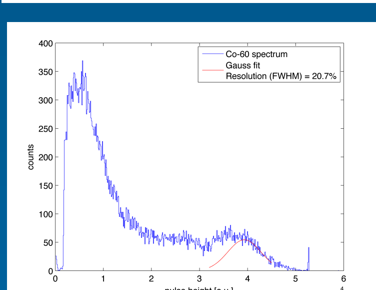
Influence of Crystal Shape

- **Cylindrical Crystal**
  - Position dependence of light collection ➔ degraded resolution
- **Cubic Crystal**
  - Less position dependence ➔ improved resolution
  - Possibly higher light collection in cryogenic detector module
  - Higher packing density possible

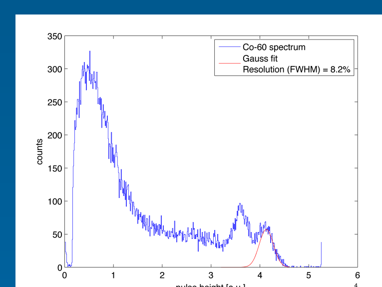


Simulated light collection





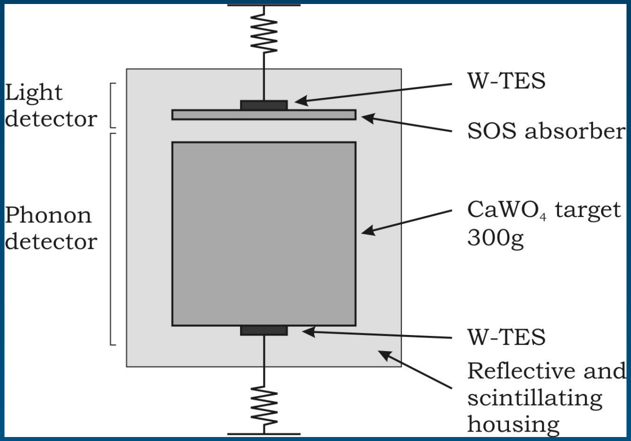
^{60}Co spectrum (1.17MeV,1.33MeV)



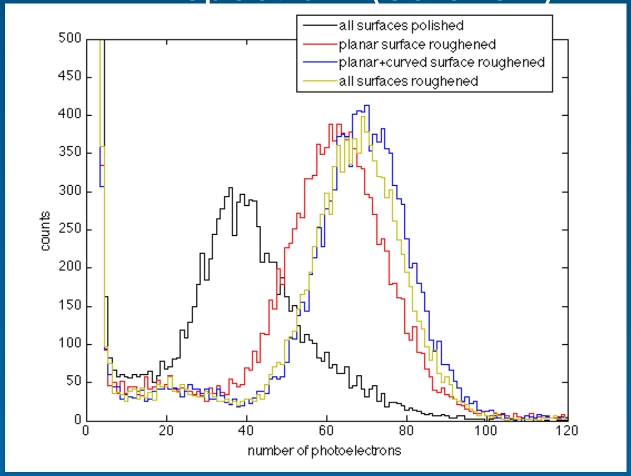
Surface Roughening

- For scintillating bolometers no optical coupling to light detector possible
  - ➢ Large fraction of light trapped in crystal (≈60% for cylindrical crystal)
- Mechanical roughening of crystal surfaces (grain size ≈9µm) reduces light trapping
  - ➢ Increases light collection efficiency
  - ➢ Reduces position dependence of light collection

CRESST-II detector scheme



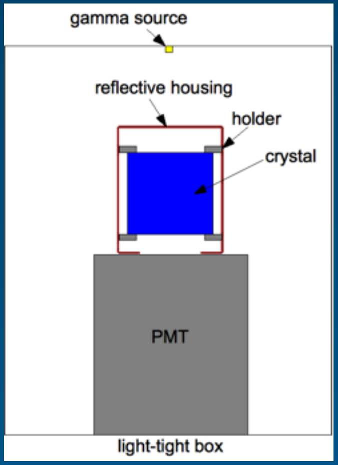
^{241}Am spectrum (59.5keV)



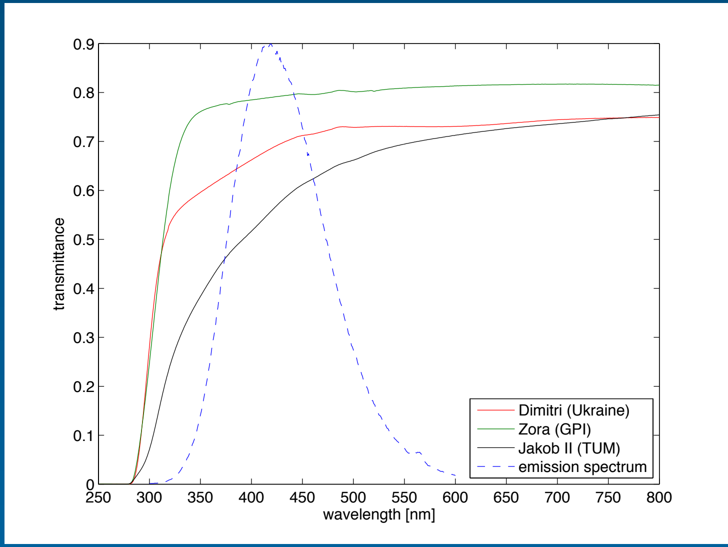
Light Yield of Crystals

- Light yield measurements at RT in comparison to reference crystal
  - ➢ Better light yield of crystals from external suppliers (especially GPI) due to smaller absorption coefficient

Setup for light yield measurements



Transmission measurement



Crystal	Light Yield	Attenuation length (@ 420nm)
Jakob II (TUM)	99%	10cm
Dimitri (Ukraine)	111%	23cm
Zora (GPI Russia)	130%	167cm

Raw Materials for Crystal Growth

- $CaWO_4$ powder prepared by solid state reaction at 1200°C from high purity $CaCO_3$ (5N) and WO_3 (4N8) powders

$$CaCO_3 + WO_3 \rightarrow CaWO_4 + CO_2$$

- Main radioactive contamination in $CaCO_3$
  - ^{226}Ra, ^{90}Sr due to similar chemical properties to Ca

Isotope	Activity (mBq/kg)		
	$CaCO_3$	**WO_3**	**$CaWO_4$**
^{232}Th	<18	<1.4	<8.9
^{238}U	<180	<468	<450
^{226}Ra	13±5	<2.2	28±6
^{40}K	<90	<20	<65
^{137}Cs	<6	<1.2	<5.5
^{60}Co	<4	<0.68	<2.6

measurements by J. Puimedon at LSC

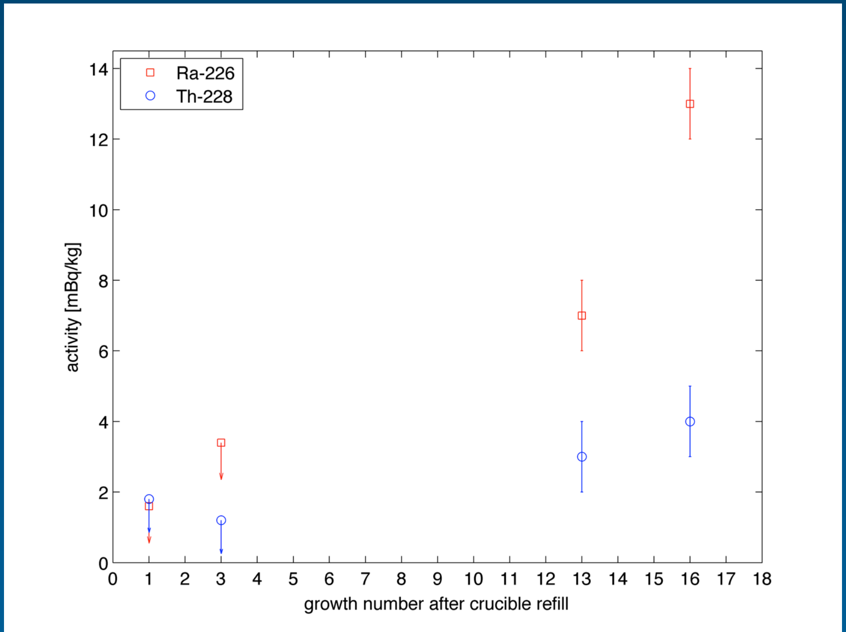
Segregation of Radioimpurities

- Impurities are usually rejected by the crystal during growth and accumulate in the melt
  - Concentration of impurities in the crystals increases with increasing growth number if residual melt is reused
  - Estimated segregation coefficients $s=c_{crys}/c_{melt}$

    $s_{Ra226}<0.12$

    $s_{U238}\approx0.3$[1]

Screening results from y-spectrometry

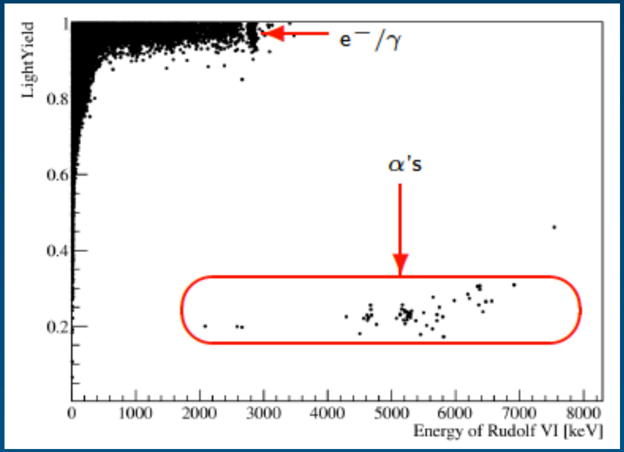


Measurements by M. Laubenstein at LNGS

[1]F.A. Danevich et al., NIM A 631, 44-53 (2011)

Alpha Activity of Crystals

- Two TUM crystals (≈300g) were operated as low-temperature detectors in CRESST test cryostat at LNGS (t_{meas}≈100-400h)
- Total alpha activity was compared to all bought crystals that were operated in last CRESST run (Diploma thesis of A. Münster)
  - **Total alpha activity of TUM crystals is in the range of radiopurest CRESST crystals (≈1-2mBq/kg)**



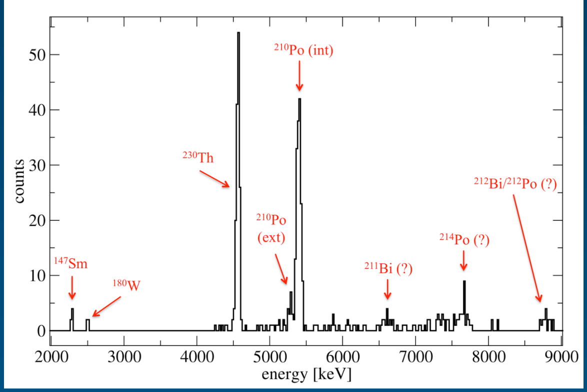
Crystal	Total alpha activity (mBq/kg)
Rudolph VI (TUM)	0.91±0.10
Wilhelm (TUM)	1.97±0.13
Daisy (GPI Russia)	2.38±0.02
Maja (GPI Russia)	66.16±0.11
SRC CARAT[1]	≈400

[1]Y. Zdesenko et al., NIM A 538, 657-667

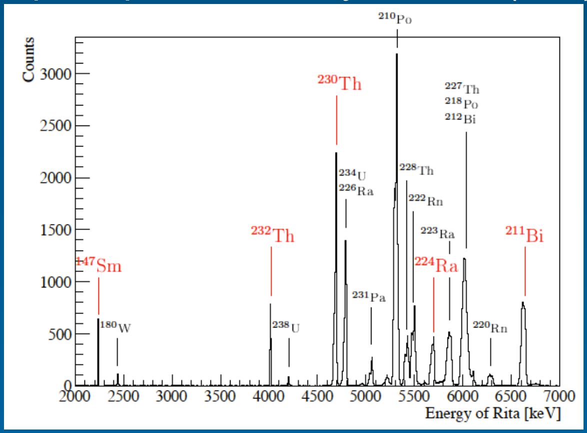
Alpha Spectra of Crystals

- Alpha spectrum from operation as low-temperature detector
- TUM crystal Wilhelm
  - Equilibrium of U-238 chain broken
  - Activity dominated by Th-230 and Po-210
- GPI crystal Rita
  - Larger contribution from rest of U-238 chain and Th-232/ U-235 chains to total activity

Alpha spectrum of crystal Wilhelm (TUM)



Alpha spectrum of crystal Rita (GPI)



Summary & Outlook

- Since 2011: Successful production of 300g $CaWO_4$ detector crystals at TUM
- 4 TUM crystals are installed in the current CRESST run (running since July 2013)
- Good radiopurity (total alpha activity: ≈1-2mBq/kg)
  - Future improvement by e.g. chemical separation of Sr, recrystallization
- Further improvement of Light Yield (transmittance) necessary
  - ➢ Vary stoichiometry of melt
- Future: Production of 1kg detector crystals for EURECA

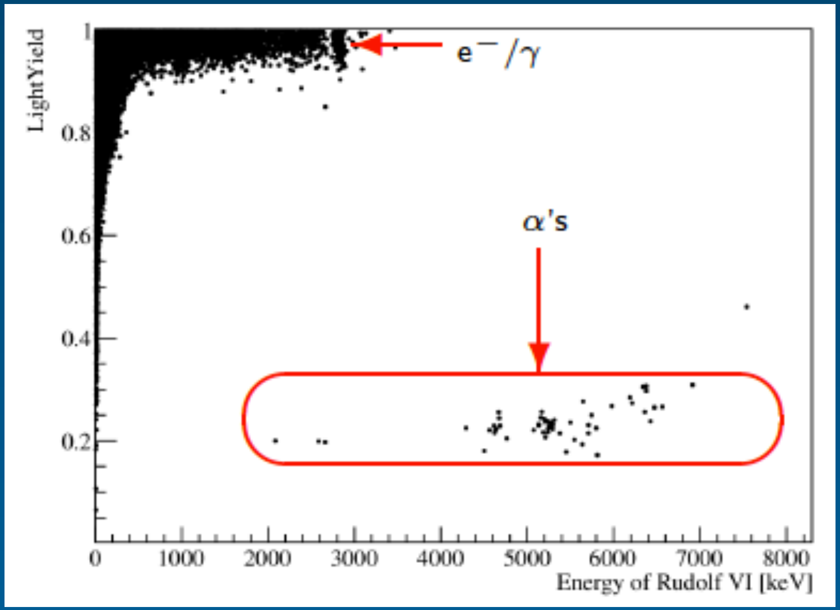
Backup Slides

Growth Flow Chart

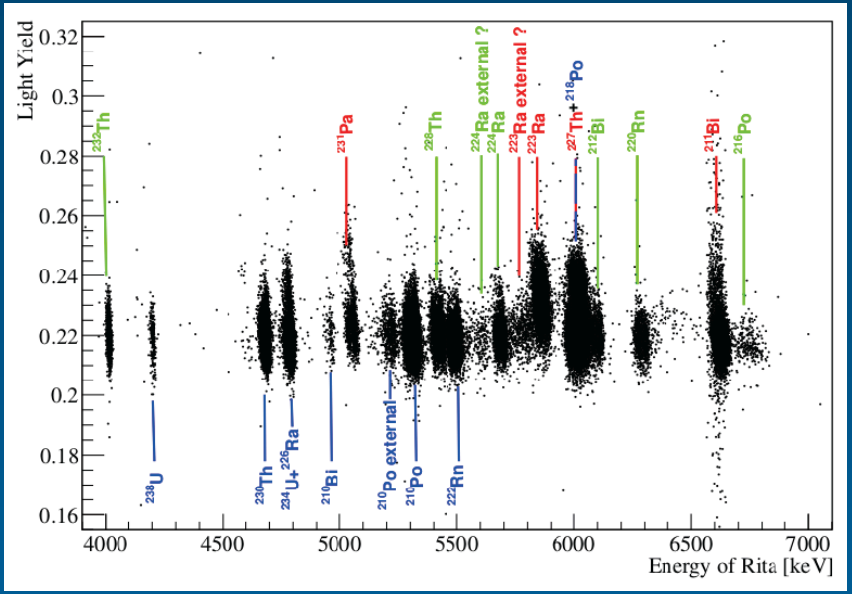
...
TUM-13
...
TUM-16
...
crucible cleaned after growth nr. 19 refilled with fresh $CaWO_4$ powder
TUM-20
...
TUM-22
...
TUM-27 (Rudolph VI)
...
crucible cleaned after growth nr. 31 refilled with parts from crystals of growths nr. 21, 29
...
TUM-40 (Wilhelm)

Light Yield Plots

Crystal Rudolph VI (TUM)



Crystal Rita (GPI Russia)



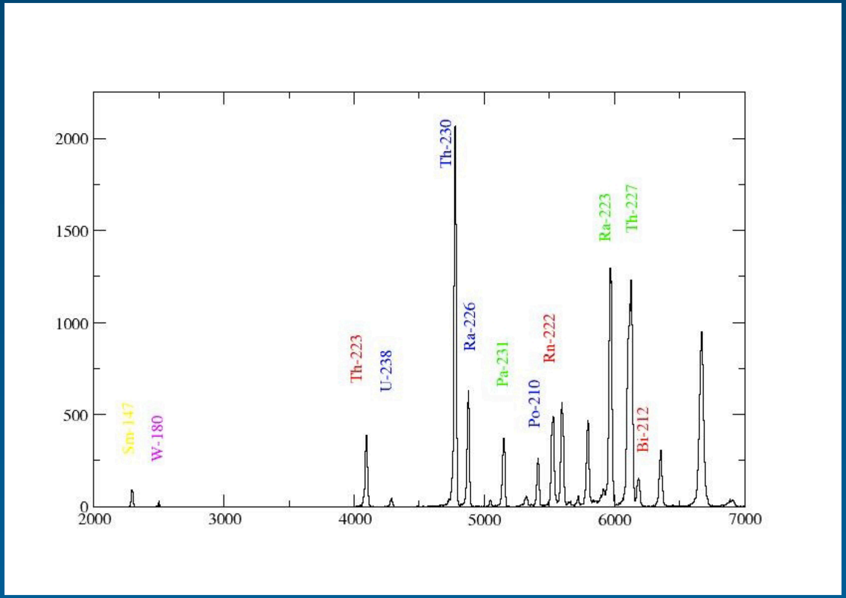
Alpha Activities in CRESST Crystals

crystal	counts	$A_{\alpha,\ \mathrm{total}}$ [mBq/kg]
Rita	76 138	10.21 ± 0.04
Daisy	17 589	2.38 ± 0.02
Maja	342 289	66.16 ± 0.11
Zora	237 245	29.68 ± 0.06
VK33	64 067	9.91 ± 0.04
Sabine	14 450	2.35 ± 0.02
Wibke	174 097	25.54 ± 0.06
Verena	65 048	11.25 ± 0.04
K07	122 732	19.45 ± 0.06

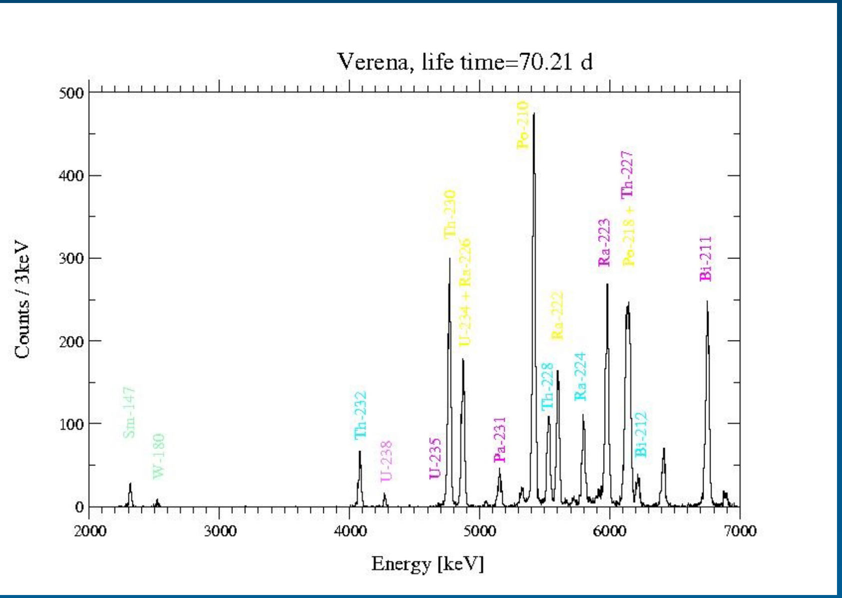
	Rita		Daisy		Maja (VK37)	
isotope	counts	A [mBq/kg]	counts	A [mBq/kg]	counts	A [mBq/kg]
^{144}Nd	< 12	< 0.002	110	0.015 ± 0.001	< 2	$< 4 \cdot 10^{-4}$
^{152}Gd	< 25	< 0.003	68	0.009 ± 0.001	< 2	$< 4 \cdot 10^{-4}$
^{147}Sm	674	0.090 ± 0.003	6288	0.850 ± 0.011	135	0.026 ± 0.002
^{180}W	232	0.031 ± 0.002	229	0.031 ± 0.002	85	0.016 ± 0.002
^{232}Th	1663	0.230 ± 0.005	284	0.038 ± 0.002	1490	0.288 ± 0.007
^{238}U	229	0.031 ± 0.002	259	0.035 ± 0.002	776	0.150 ± 0.005
^{230}Th	8036	1.077 ± 0.012	589	0.080 ± 0.003	111 782	21.605 ± 0.065
^{234}U	5366	0.719 ± 0.010	725	0.098 ± 0.004		
^{226}Ra						
^{210m}Bi	126	0.017 ± 0.002	51	0.007 ± 0.001	2015	0.390 ± 0.009
^{231}Pa	1158	0.155 ± 0.005	78	0.011 ± 0.001		
^{210}Po ext	698	0.094 ± 0.004	504	0.068 ± 0.003	111 611	21.572 ± 0.065
^{210}Po int	18 690	2.506 ± 0.018	809	0.109 ± 0.004		
^{228}Th	2995	0.402 ± 0.007	306	0.041 ± 0.002		
^{222}Rn	5159	0.692 ± 0.010	468	0.063 ± 0.003		
^{224}Ra	2900	0.388 ± 0.007	270	0.037 ± 0.002	7667	1.482 ± 0.017
^{223}Ra	4924	0.660 ± 0.009	1498	0.203 ± 0.005		
^{218}Po	12 298	1.648 ± 0.015	2254	0.305 ± 0.006	94 440	18.253 ± 0.059
^{227}Th						
^{212}Bi (36%)	932	0.125 ± 0.004				
^{220}Rn	934	0.125 ± 0.004	123	0.017 ± 0.001	1037	0.200 ± 0.006
^{211}Bi	7325	0.982 ± 0.011	1665	0.225 ± 0.006	4763	0.921 ± 0.013
^{216}Po	228	0.031 ± 0.002	< 10	< 0.001		

Alpha Spectra of CRESST Crystals

Crystal Zora



Crystal Verena



Radiopurity of Crystals from γ-Spectrometry

- Offcut of raw Ingot screened with HPGe detector
  - TUM crystal shows small ^{137}Cs contamination (≈1.6mBq/kg)
  - Lower ^{226}Ra activity compared to CARAT crystal

Isotope	Activity (mBq/kg)		
	TUM[2]	GPI Russia[2]	SRC “CARAT”[1]
^{228}Th	<1.8	<4	0.6±0.2
^{228}Ac	<1.4	-	-
^{234m}Pa	<47	<90	14.0±0.5
^{226}Ra	<1.6	<6	5.6±0.5
^{40}K	<8.7	<17	<12
^{137}Cs	1.6±0.5	<1	<0.8
^{60}Co	<0.3	<2	-

[1]Y. Zdesenko et al., NIM A 538, 657-667
[2]Measurements by M. Laubenstein at LNGS

Natural Decay Chains

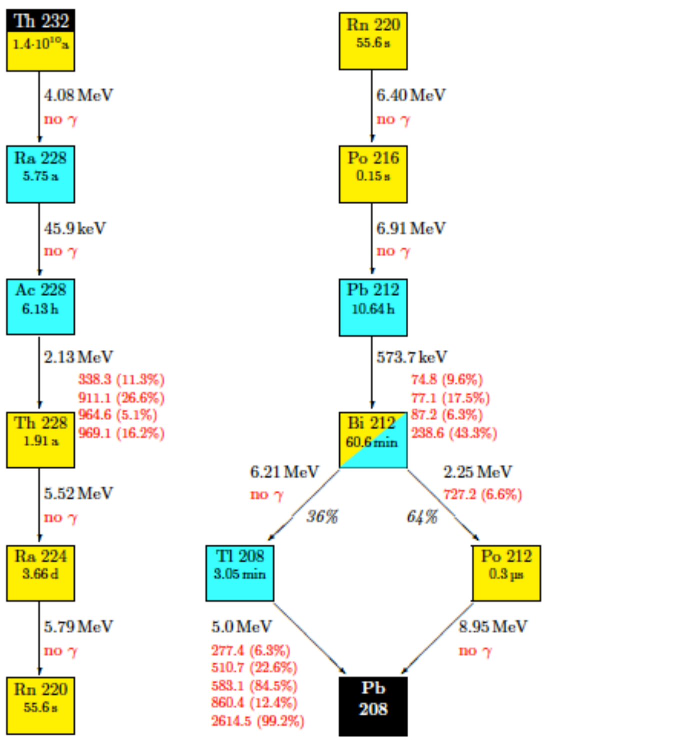


Figure A.2: *Full decay chain of ^{232}Th. All branchings with a branching ratio less than 1% are neglected. The half-lives of the radioisotopes are given, as well as the Q-values of the decays.*

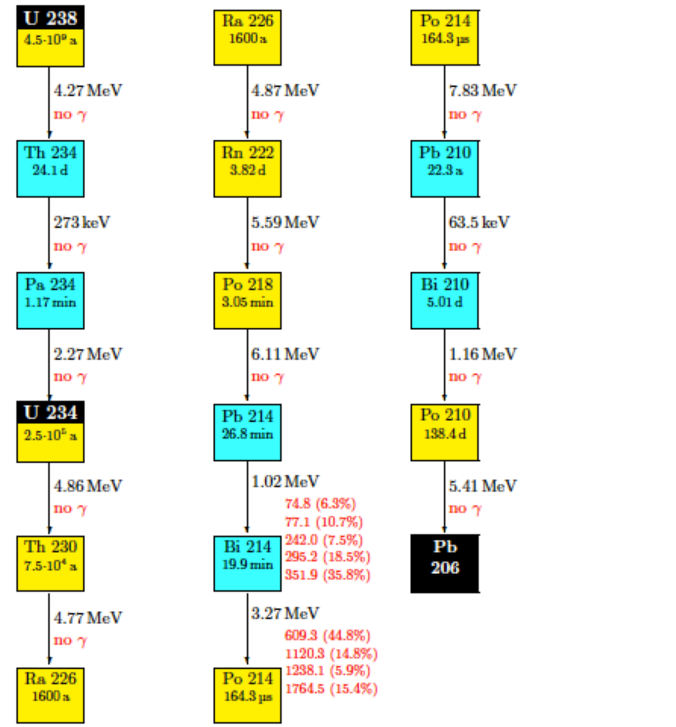
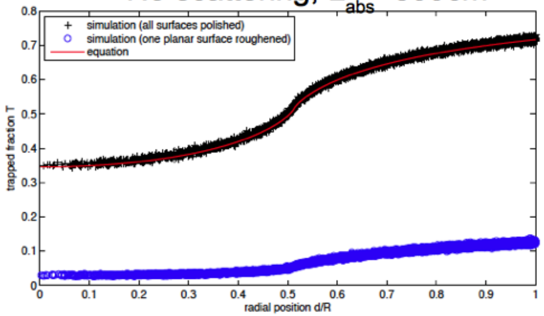


Figure A.1: *Full decay chain of ^{238}U. All branchings with a branching ratio less than 1% are neglected. The half-lives of the radioisotopes are given, as well as the Q-values of the decays. Note that in the decay of ^{234}Th the isotope ^{234}Pa is populated in an isomeric state, which does not de-excite to the ground state, but undergoes a β^--decay into ^{234}U in 99.84% of all cases.*

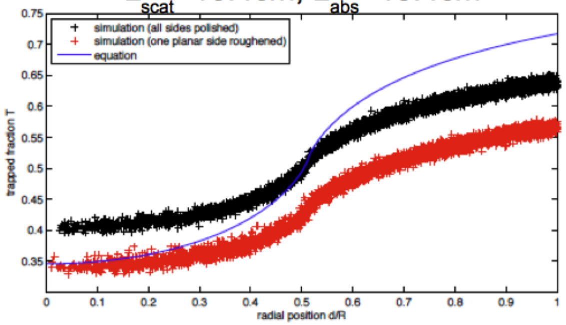
Light Trapping in Cylinder

Light trapping in cylindrical crystals

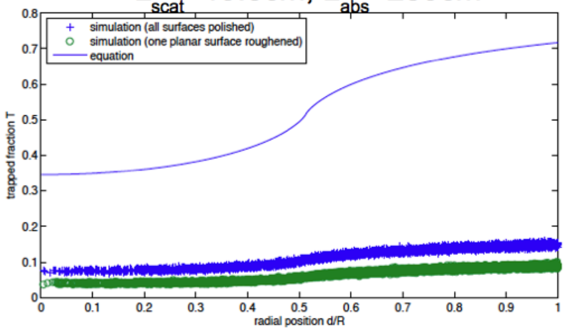
No scattering, L_{abs}=500cm



L_{scat}=16.4cm, L_{abs}=15.4cm



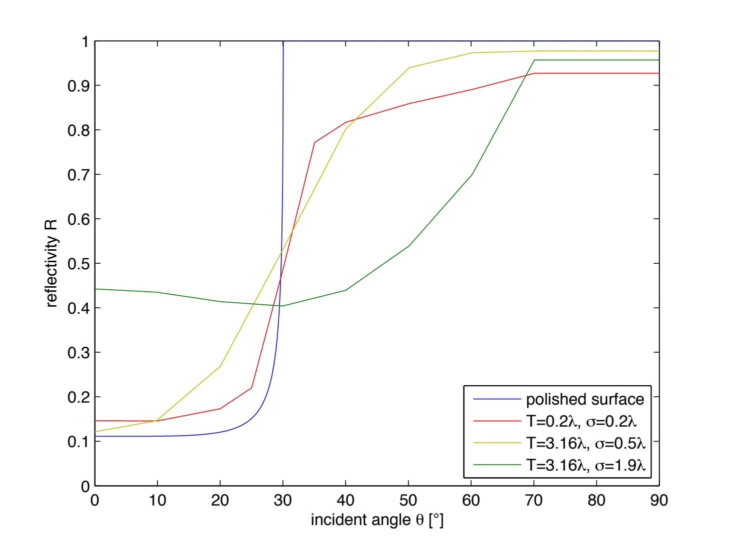
L_{scat}=19.3cm, L_{abs}=250cm

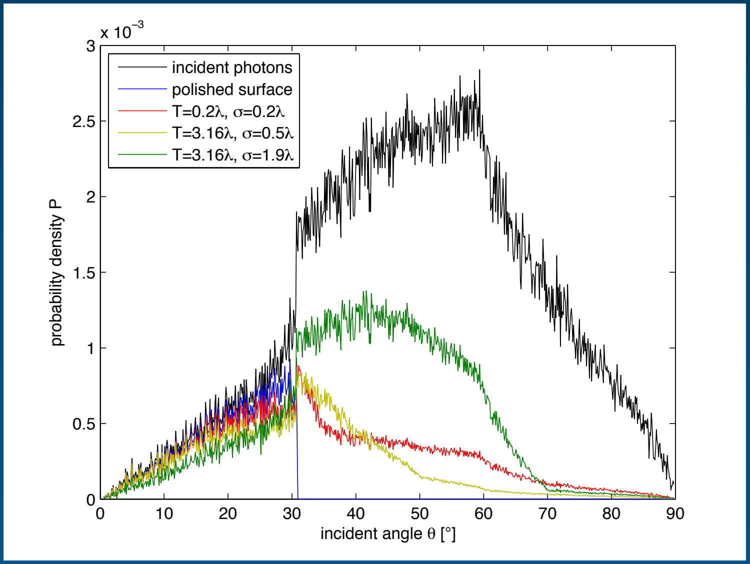


- Fraction T of trapped light depends on radial position of scintillation event (→ σ_{pos}) and increases with distance from cylinder axis[1]
- Values of T, σ_{pos} and profit of roughening depends on absorption and scattering lengths → preferable: high L_{abs}, small L_{scat}

[1] C. Carrier and R. Lecomte, Nucl. Instrum. Methods Phys. Res. A 278, 622 (1989).

Influence of Surface Roughening on Transmittance





- Critical angle for total internal reflection $\theta_c \approx 30°$
- Roughness with correlation length T and height distribution σ greater than wavelength λ
  - ➢ Prevention of total internal reflection for incident angles $>\theta_c$

M. Nieto-Vesperinas and J. A. Sanchez-Gil, J. Opt. Soc. Am. A 9, 424 (1992).

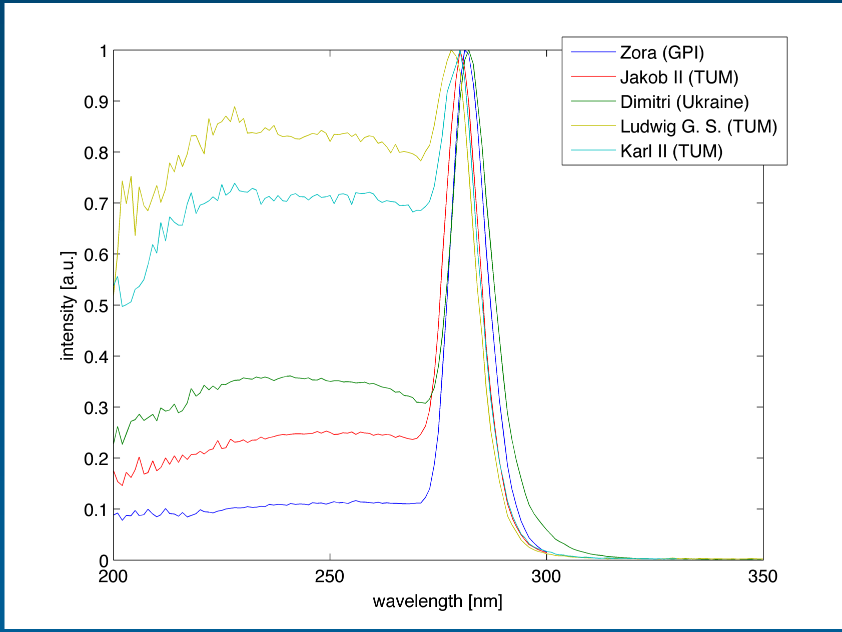
Influence of Growth Atmosphere

Crystal grown under pure N_2 atmosphere

Crystal grown under 99% Ar, 1% O_2 atmosphere

Luminescence Spectra

Excitation spectra



Emission spectra