

TeO₂ scintillating crystals growth and properties

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Abstract

The advantages of using hybrid detectors (scintillating bolometers) in rare event searches like Double-Beta Decay or Dark Matter interaction are discussed.

The possibility to produce scintillating TeO₂ crystals is emphasized and the results of optical transmission and X-ray excited steady-state luminescence measurements performed on pure and doped TeO₂ crystals are presented.

CUORE experiment



CUORE: Cryogenic Underground Observatory for Rare Events

At extremely low temperatures (~10 mK), even small energy release in a crystal results into a measurable temperature rise (heat capacity ~ T T_D where T_D is the Debye temperature)

CUORE needs 4 x 700g TeO₂ crystals

Crystals of TeO₂ may act both as a detector and source

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goal: achieve a background rate in the range 0.001 to 0.01 counts/keV/kg at the 0ννββ-decay transition energy of ¹²⁵Te (2.038 MeV).

scintillation may help to discriminate the type of interacting quantum with an immediate effect on background reduction

What is known on scintillation in TeO₂

• L_{scint} = 0.67% BGO

• no indication on spectrum or decay characteristics

• P. de Marcellis, INFN LTD, Genova, 7-11 July 2001

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paratellurite

Characteristics value

Chemical Formula TeO₂

Crystallographic System Tetragonal

Crystal Class 4

Crystallographic Axes a, b, c

Crystallographic Angles α, β, γ

Crystallographic Density ρ

Crystallographic Refractive Index n_x, n_y, n_z

Crystallographic Thermal Expansion Coefficient α_x, α_y, α_z

Crystallographic Thermal Conductivity κ_x, κ_y, κ_z

Crystallographic Thermal Stability

Crystallographic Mechanical Properties

Crystallographic Optical Properties

Crystallographic Electrical Properties

Crystallographic Magnetic Properties

Crystallographic Acoustic Properties

Crystallographic Piezoelectric Properties

Crystallographic Pyroelectric Properties

Crystallographic Ferroelectric Properties

Crystallographic Superconducting Properties

Crystallographic Semiconducting Properties

Crystallographic Insulating Properties

Crystallographic Dielectric Properties

Crystallographic Piezoresistive Properties

Crystallographic Thermoelectric Properties

Crystallographic Magnetoelectric Properties

Crystallographic Multiferroic Properties

Crystallographic Topological Properties

Crystallographic Quantum Properties

Crystallographic Nanoscale Properties

Crystallographic Microscale Properties

Crystallographic Mesoscale Properties

Crystallographic Macroscale Properties

Crystallographic Geoscale Properties

Crystallographic Cosmological Properties

Crystallographic Astrophysical Properties

Crystallographic Planetary Properties

Crystallographic Interplanetary Properties

Crystallographic Interstellar Properties

Crystallographic Extraterrestrial Properties

Crystallographic Universal Properties

Crystallographic Multiversal Properties

Crystallographic Hyperuniversal Properties

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Crystal

TeO₂ scintillating crystals growth and properties

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CUORE experiment



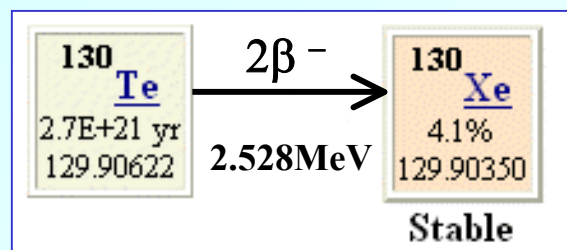
- *observation and measurement of the neutrinoless double-beta decay*

$$(Z, A) \rightarrow (Z + 2, A) + 2e^{-}$$

- *unique way to look for neutrino masses of the Majorana type*

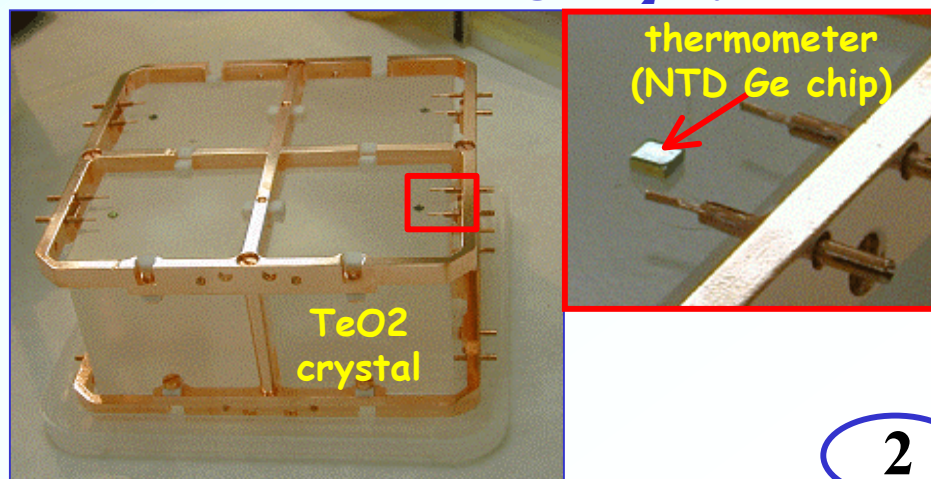
Cryogenic Underground Observatory for Rare Events

At extremely low temperatures ($\sim 7\text{-}10\text{ mK}$), even small energy release in a crystal results into a measurable temperature rise (heat capacity $\sim T/T_D^3$ where T_D is the Debye temperature)



Crystals of TeO_2 may act both as a detector and source

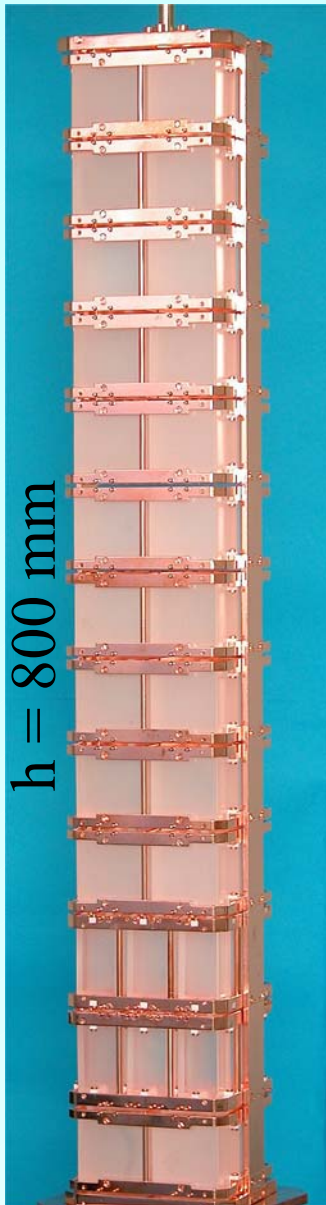
CUORE module: 4 x 760g TeO_2 crystals



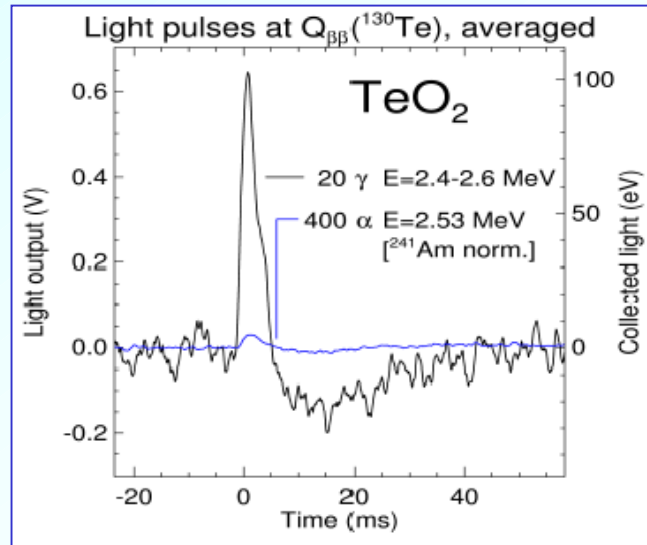
CUORE detector will consist of an array of 988 TeO_2 bolometers arranged in 19 towers of 52 crystals each.

goal: achieve a background rate in the range 0.001 to 0.01 counts/keV/kg/y at the $0\nu\beta\beta$ -decay transition energy of ^{130}Te (2.528 MeV).

scintillation may help to discriminate the type of interacting quantum with an immediate effect on background reduction



What is known on scintillation in TeO_2



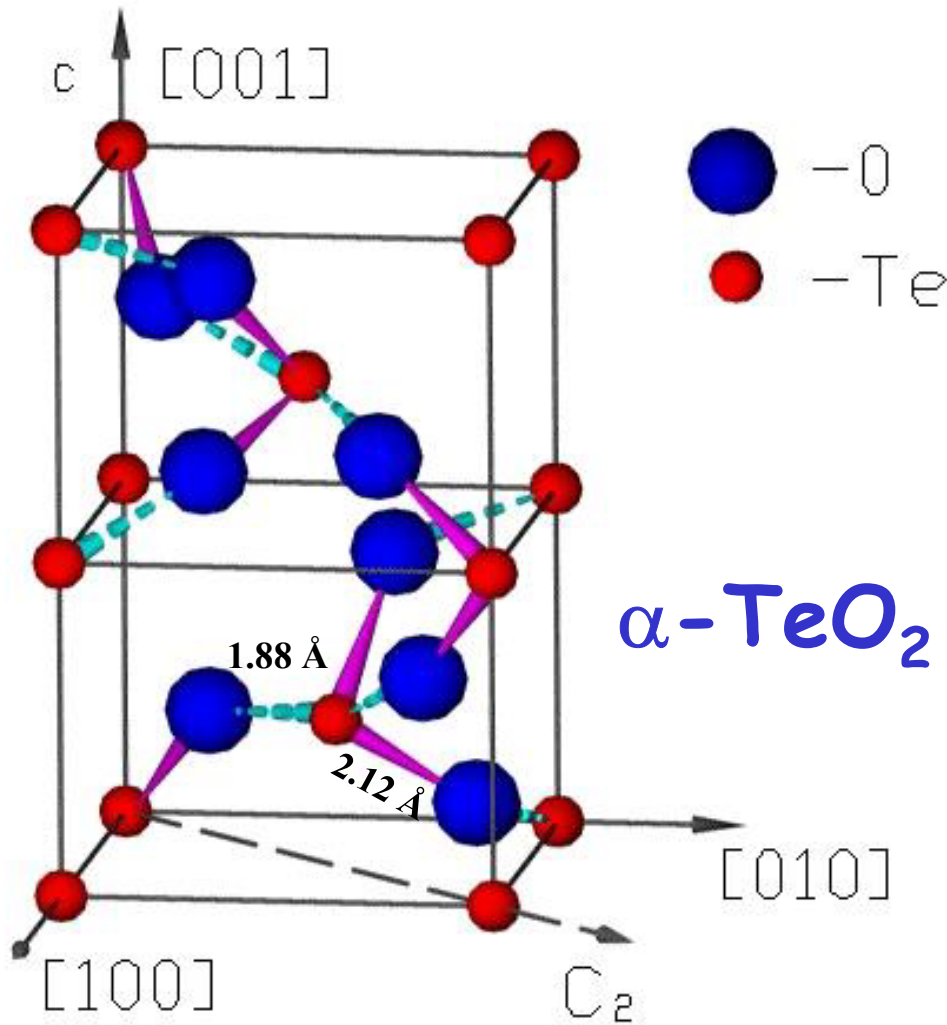
- **LY=0.67% BGO**
- **no indication on spectrum or decay characteristics**

P. de Marcillac

*10th LTD,
Genoa,*

7-11 July 2003

paratellurite



Characteristic	value
Chemical Formula	TeO_2
Molecular Weight	159.61
Crystal Class	Tetragonal
Density, g/cm^3 at 293 K	6
Melting Temperature (K)	1006
Hardness (Mohs)	4
Solubility in water	None
Color	Clear
Transmittance Range, μm	0.33-5.0
Refractive index ($\lambda=500\text{nm}$)	$n_o=2.3194$
	$n_e=2.4829$
Melting point ($^{\circ}\text{C}$)	733
Thermal Expansion at 293K, 1/K	
normal to $\langle 001 \rangle$	19.5×10^{-6}
parallel to $\langle 001 \rangle$	6.10×10^{-6}
Thermal Conductivity, mW/cm K	30
Mechanical Impedance, kg/m sec	3.6×10^6

- *Bridgman grown crystals are more stressed than Czochralski ones*
- *annealing at about 550°C helps in removing the residual stresses*
- *lateral faces parallel to 110 and $1\text{-}10$ are preferred.*
- *lateral faces parallel to the 100, 010 cleavage should be avoided*
- *doping extremely difficult due to asymmetric Te – O covalent bonds*

Activator selection criteria

general

- luminescence properties of the free ion
- incorporation to the host lattice

particular

- only dopants having stable isotopes are acceptable
- unchanged thermal characteristics of doped crystals
(*incorporation of uneven ions, avoided*)

no restriction on the emission spectrum and/or on the decay time of the activator

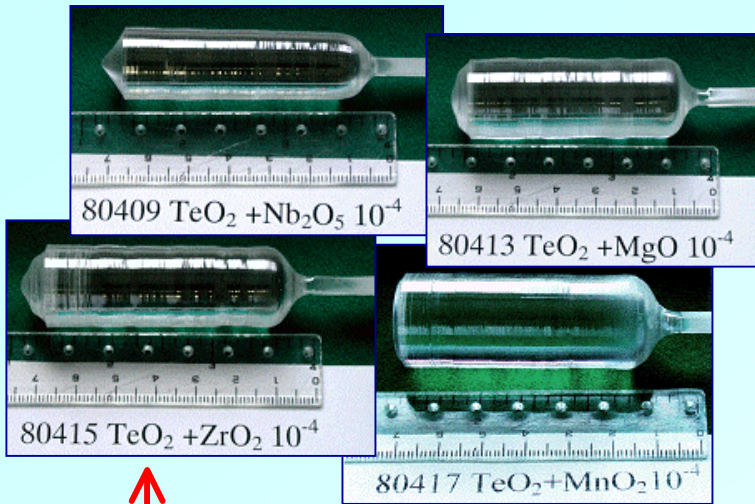
		At. Nb.	Electron config. of element	Oxi. No.	Coo. No.	ELAff (eV)	Fermi (eV)	Work func. (eV)	IonicR (Å)	El. Neg. (V)	Chem. Hard.	Free ion emission (nm)	Stable isot. (%)	Dev. IonR (%)
✕	Te	52	[Kr] 4d ¹⁰ 5s ² 5p ⁴	4	4	6.57	8.77	10.97	0.66	1.71	0.68		33.606	0.00
1	Mg	12	[Ne] 3s ²	2	5				0.66			200 - 9900	100.000	0.00
2	Mn	25	[Ar] 3d ⁵ 4s ²	2	6				0.66			204 - 350	100.000	0.00
	Mn	25	[Ar] 3d ⁵ 4s ²	2	4				0.67	1.44	0.75		100.000	1.52
	Mn	25	[Ar] 3d ⁵ 4s ²	3	6	5.86	8.14	10.41	0.645	1.62	0.69	202 - 627	100.000	-2.27
3	Zr	40	[Kr] 4d ¹⁰ 5s ¹	4	5				0.66			479 - 634	97.200	0.00
4	Nb	41	[Kr] 4d ⁴ 5s ¹	4	6	6.34	8.66	10.97	0.68			203 - 225	100.000	3.03
	Nb	41	[Kr] 4d ⁴ 5s ¹	5	6				0.64			-	100.000	-3.03
	Nb	41	[Kr] 4d ⁴ 5s ¹	5	7				0.69				100.000	4.55

In order to avoid constitutional supercooling, doped crystals have to be grown under high thermal gradient with growth conditions similar to that of the high temperature solutions growth.

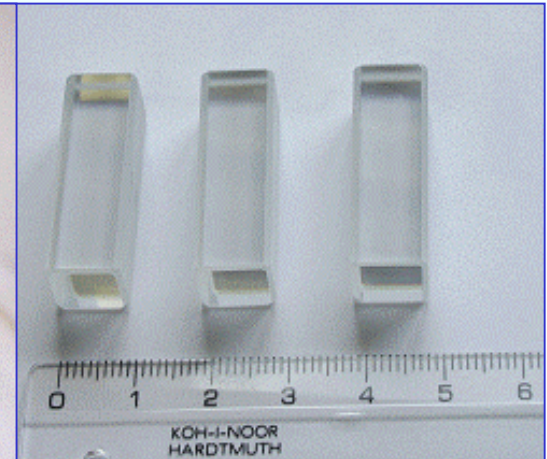
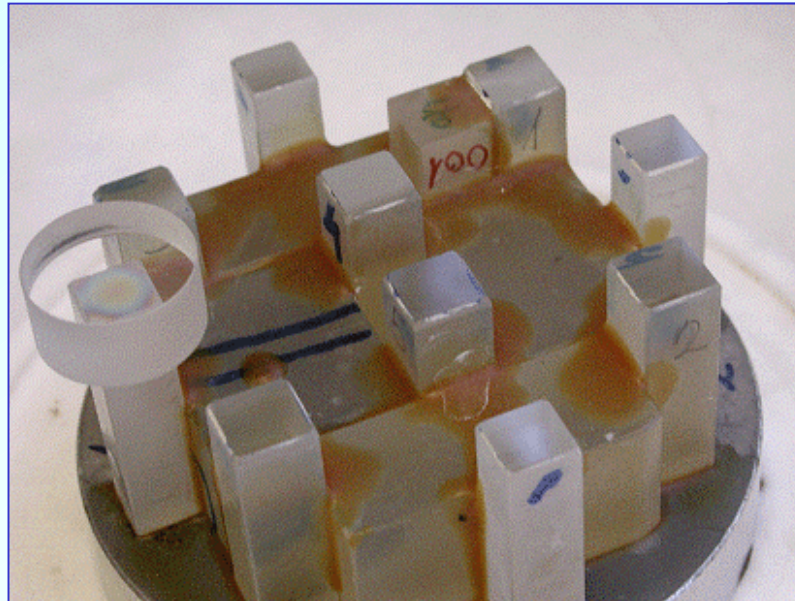
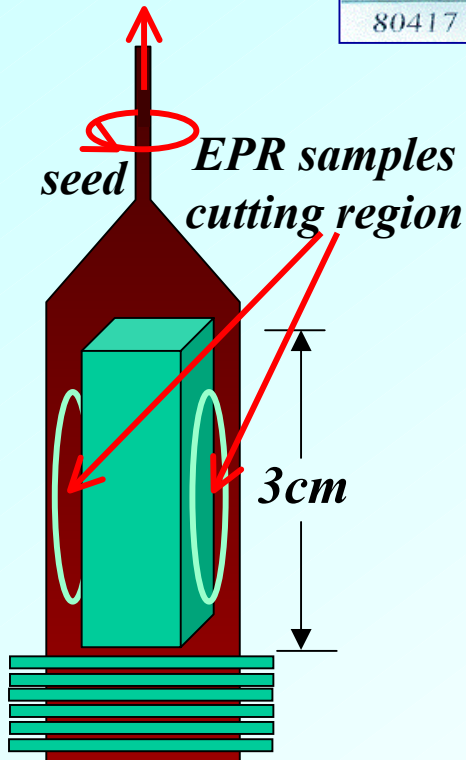
Growth parameters:

- axial gradient: 15-20 °C/cm
- pulling rate: 0.65-0.70 mm/h
- seed rotation: 26-28 rpm
- starting charge: 180g
- yield: 50-67 wt%

Sample preparation



- TeO_2 cleaves along the (100) and (010) planes. The 45 degree orientations of the cleavage planes at the edges favor the nucleation of minor cracks at the edges.
- Samples glued with melted wax ($\sim 70^\circ\text{C}$) were polished simultaneously with the help of “phantoms” of the same type of the crystal.



Work plan

1. low temperature transmission

2. steady state scintillation and TSL

3. low temperature photoluminescence

4. check for traps existence at very low temperatures

5. heat capacity

test the successful doping of TeO₂, see if the scintillation characteristics may be improved

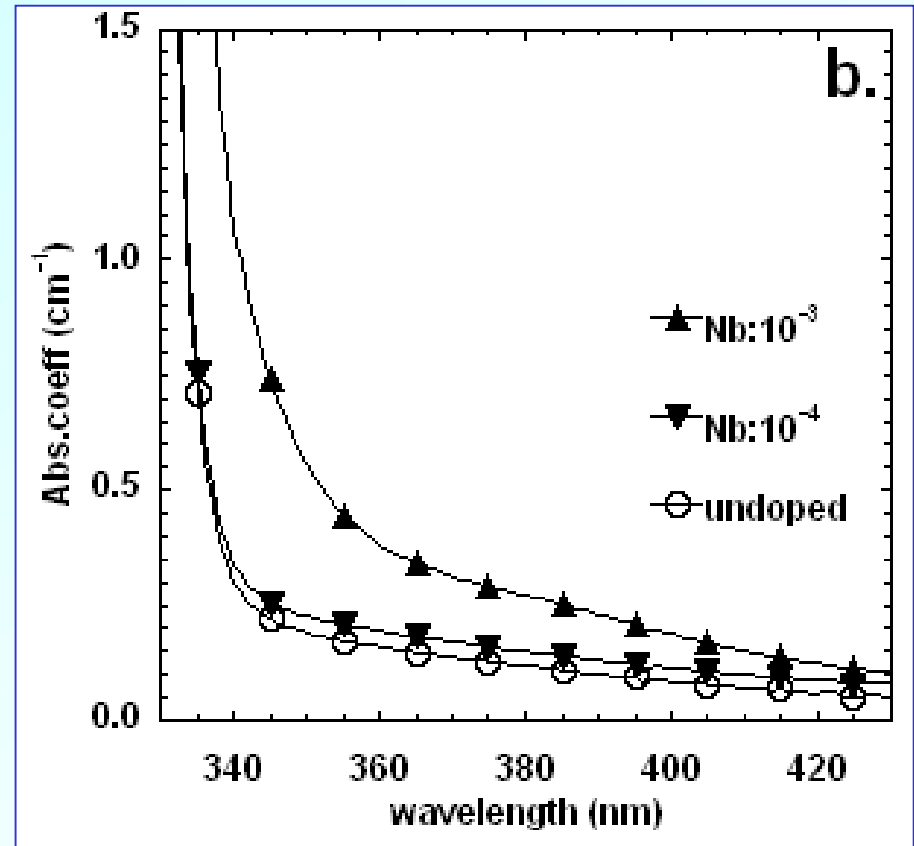
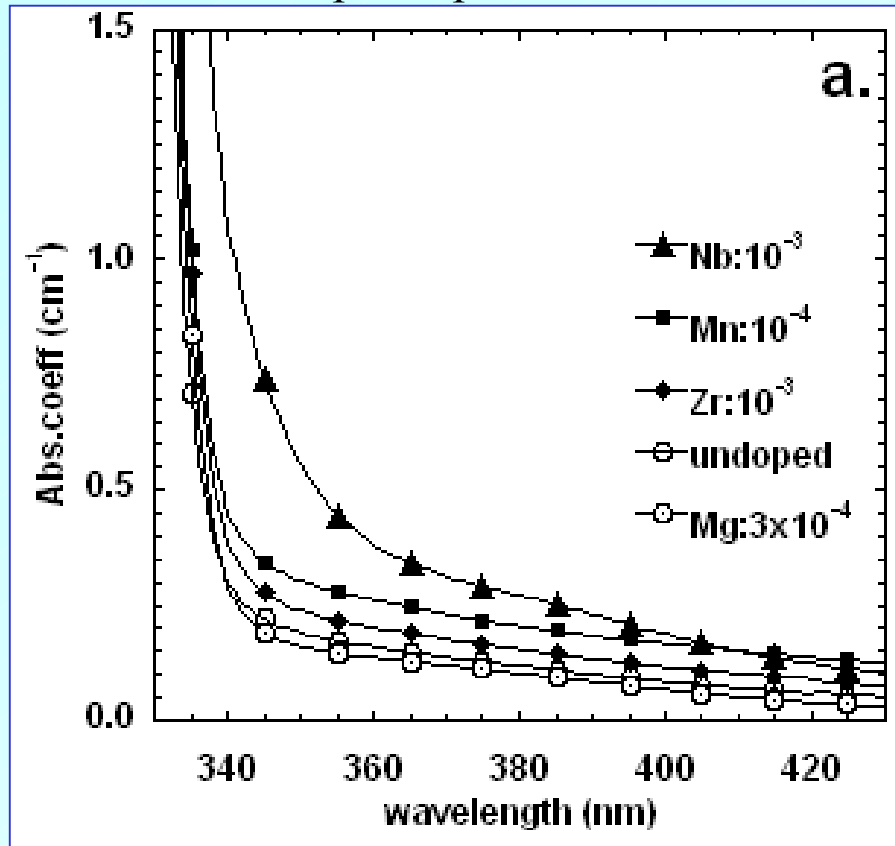
find the nature of luminescence in pure TeO₂ ; study the TeO₂:X luminescence characteristics

further study of possible improvement of TeO₂ scintillation characteristics

test the influence of doping on TeO₂ thermal characteristics; possibly find correlation between thermal characteristics at low temperatures (mK) and other characteristics measurable at higher temperatures (LN?)

Transmission at RT

JASCO V550 spectrophotometer



a. Absorption spectra of undoped, Mg, Mn, Nb and Zr doped TeO_2 crystal

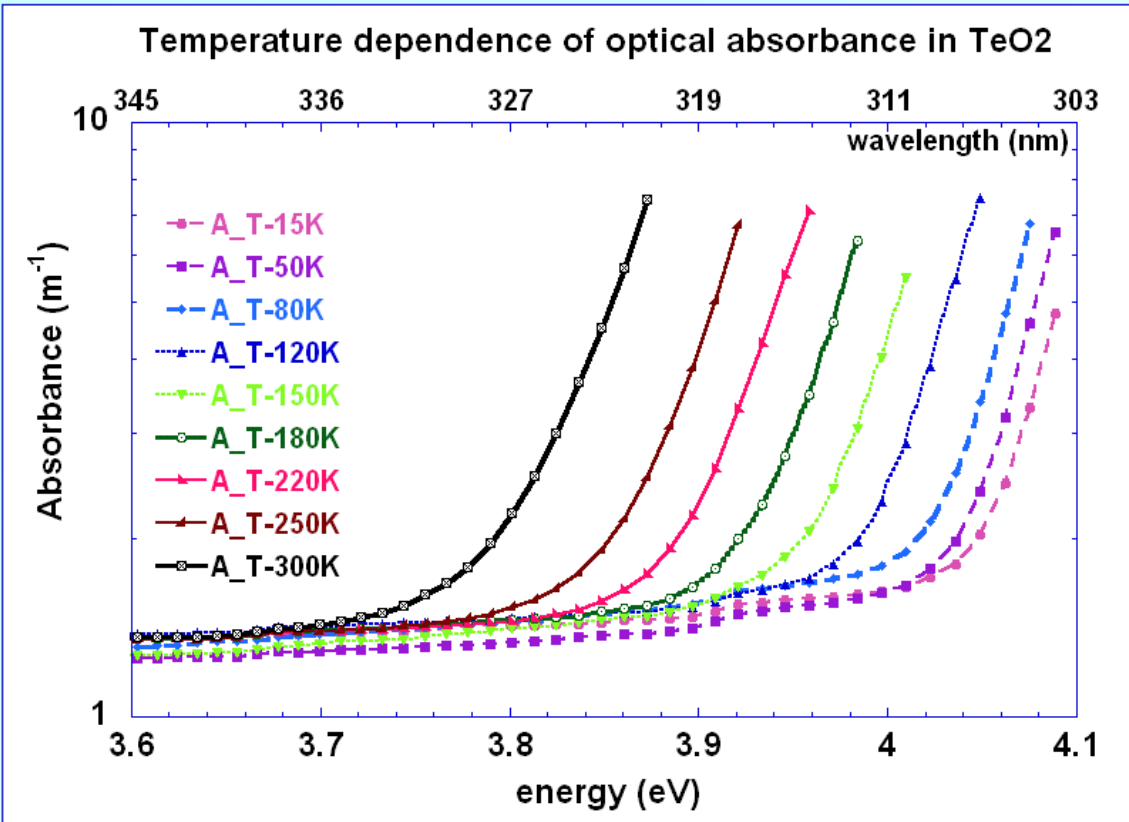
b. Absorption spectra of undoped, 10^{-4} and 10^{-3} mol/mol Nb doped TeO_2 crystals

- *Incorporation of Nb, Mn, Zr dopants is obvious.*
- *Mg incorporation is below precision limit.*

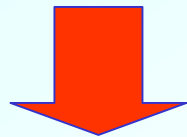
Transmission at low temperatures

Perkin Elmer *Lambda900*

Leibold *RD10-320* (Tmin=12K)



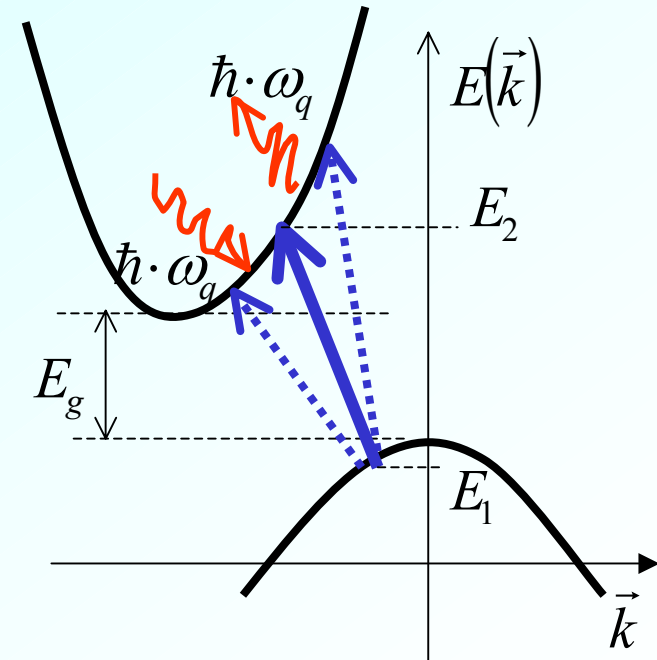
Typical behavior for indirect band transition



data analysis on the way

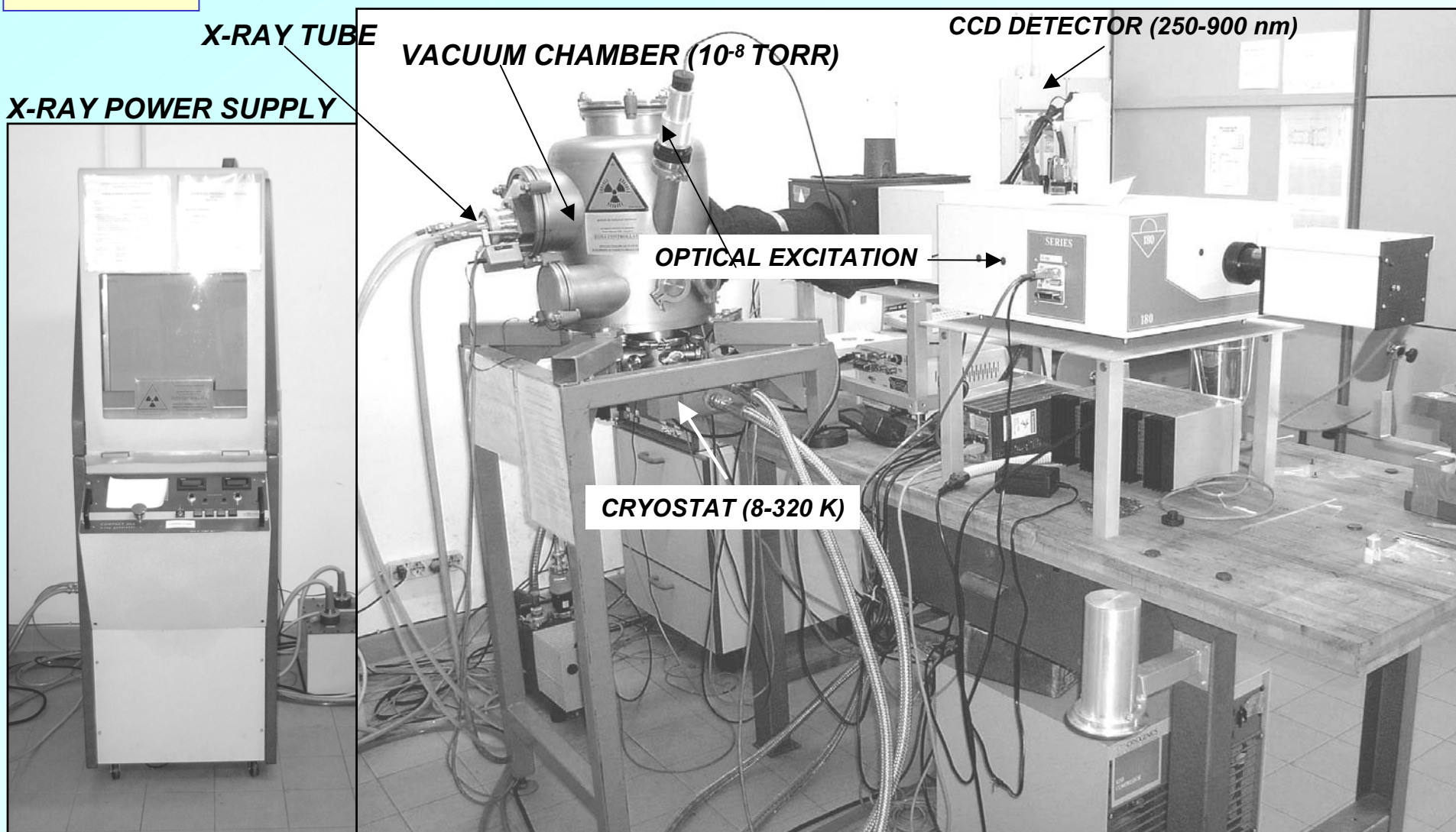
indirect transitions:

$$\vec{k}_1 \neq \vec{k}_2; E_2 - E_1 = \hbar \cdot \omega \pm \hbar \cdot \omega_q$$

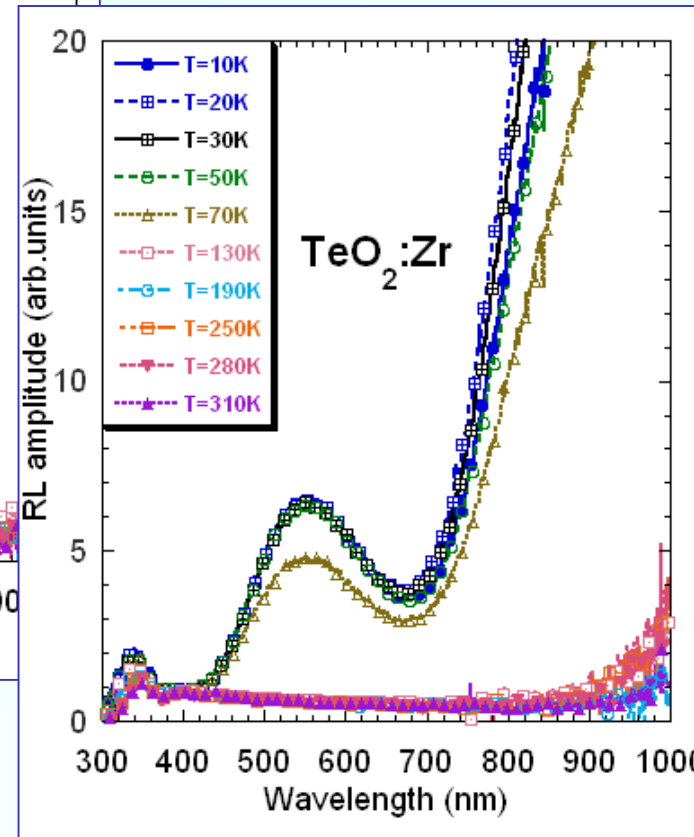
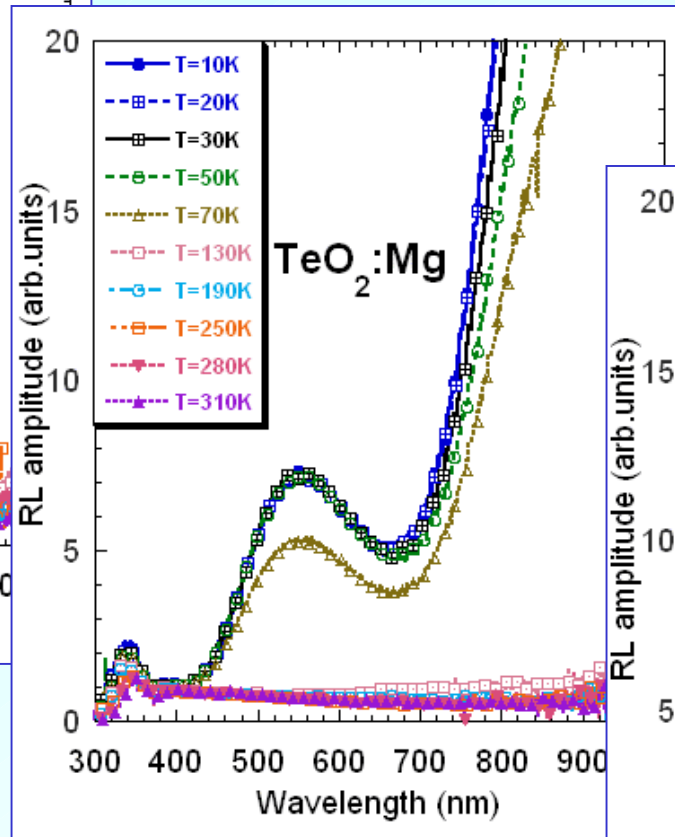
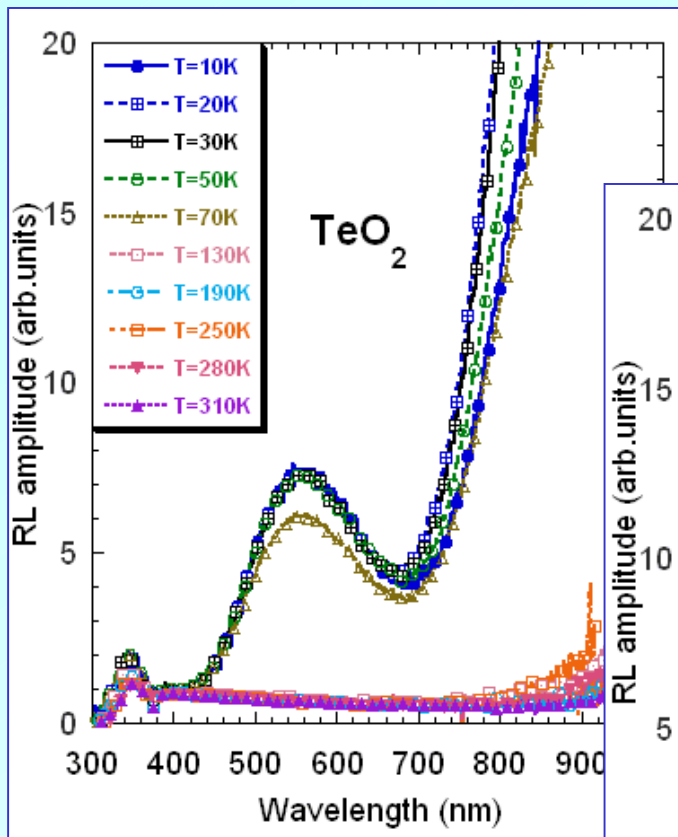


$$\alpha = \frac{const}{\hbar \cdot \omega} \cdot \left(\frac{(\hbar \cdot \omega - E_g + \hbar \cdot \omega_q)^2}{e^{\frac{\hbar \omega_q}{kT}} - 1} + \frac{(\hbar \cdot \omega - E_g - \hbar \cdot \omega_q)^2}{1 - e^{\frac{\hbar \omega_q}{kT}}} \right)$$

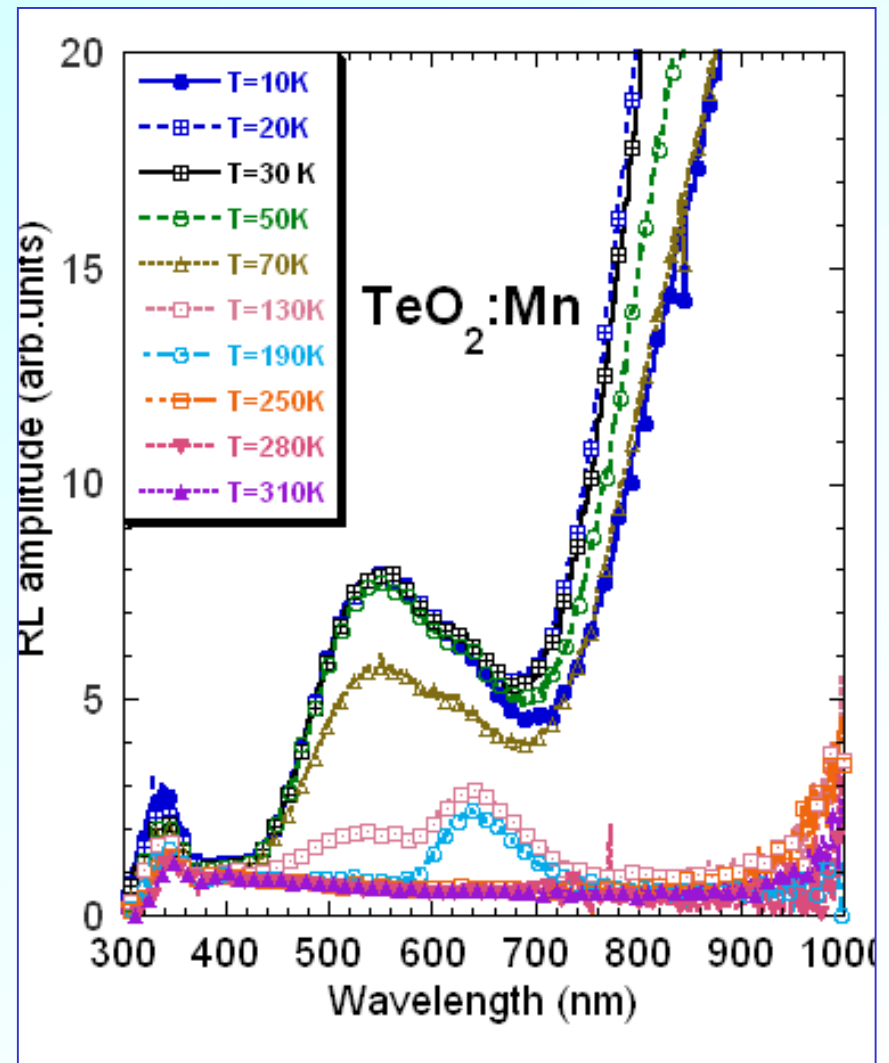
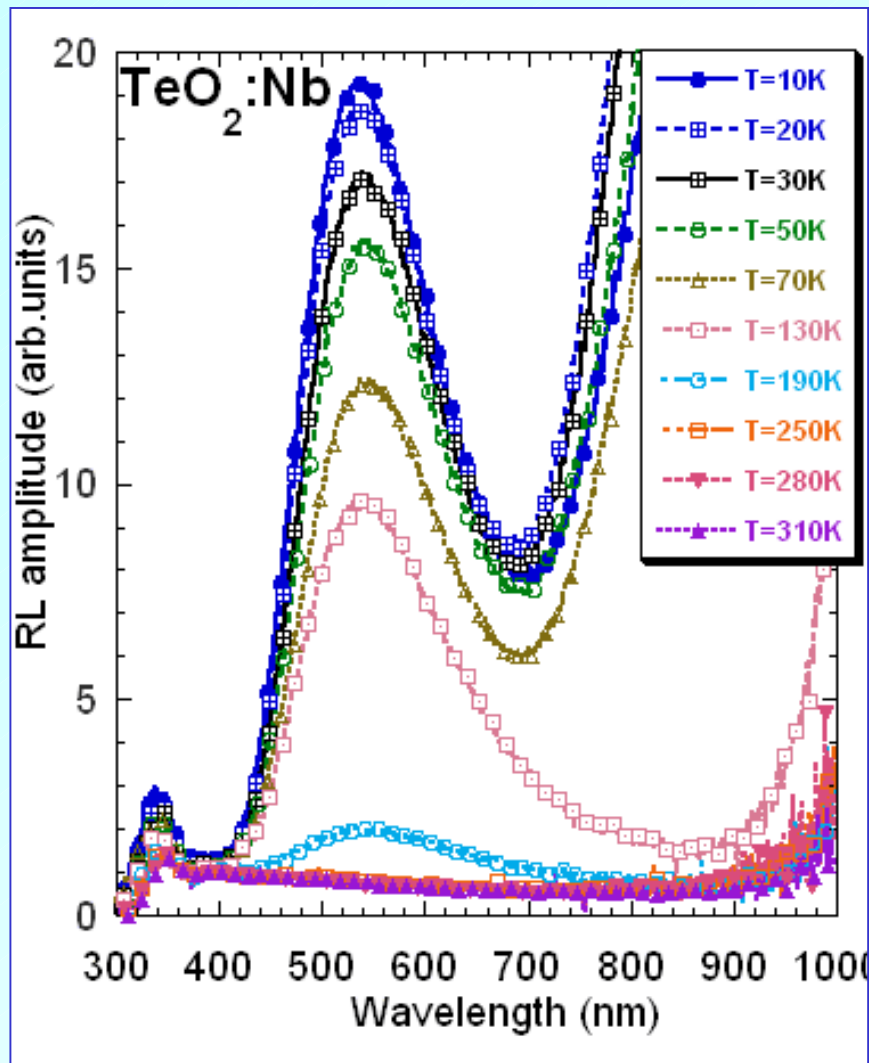
RADIO-PHOTO-THERMO LUMINESCENCE SET-UP



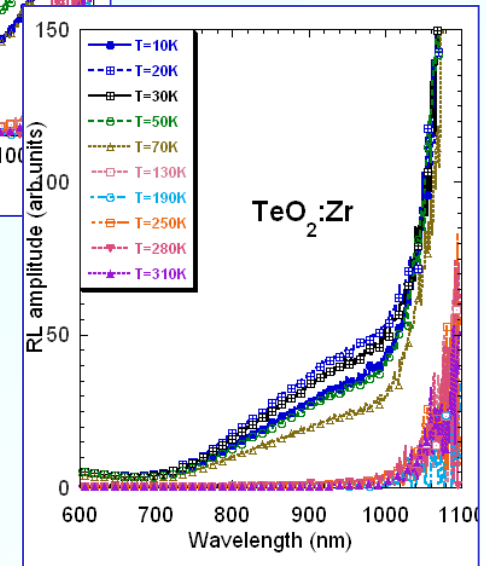
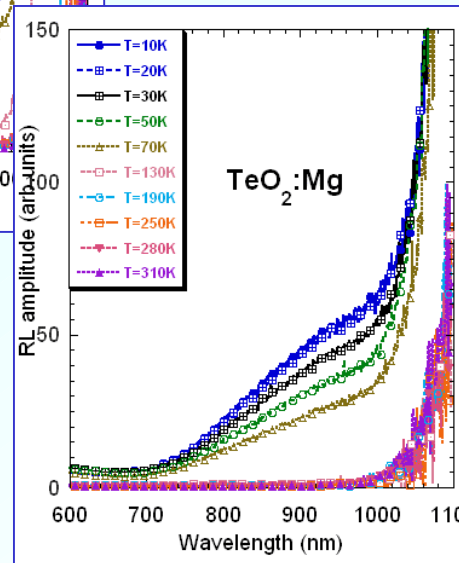
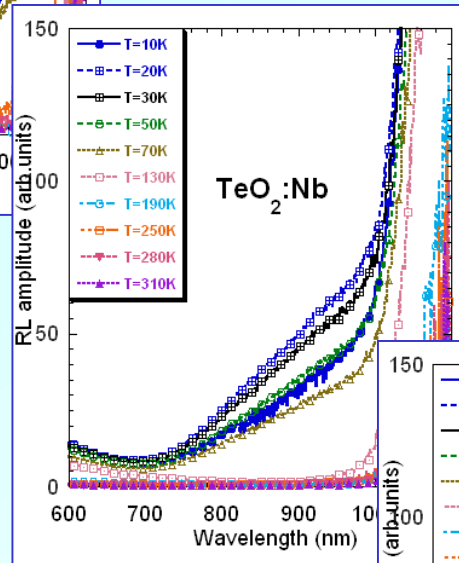
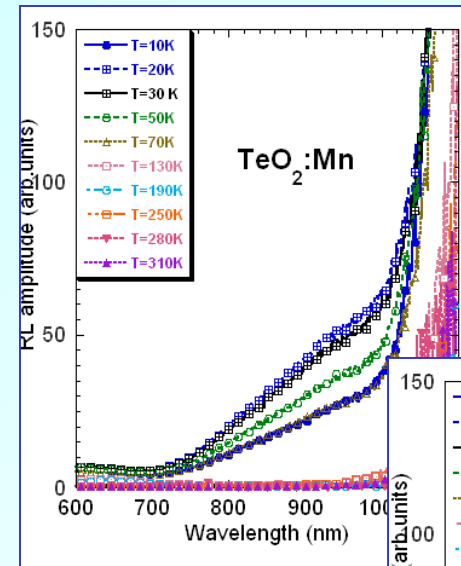
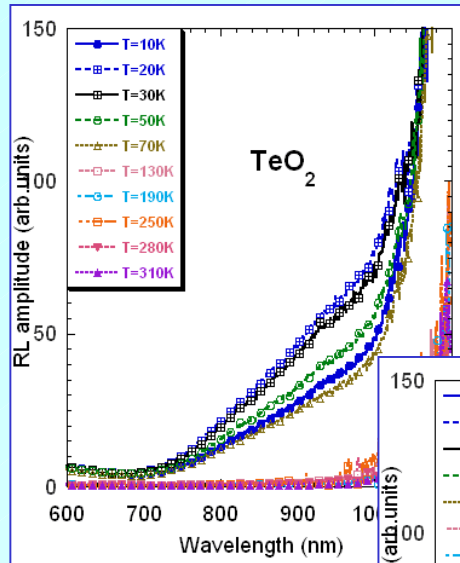
X ray excited steady-state luminescence



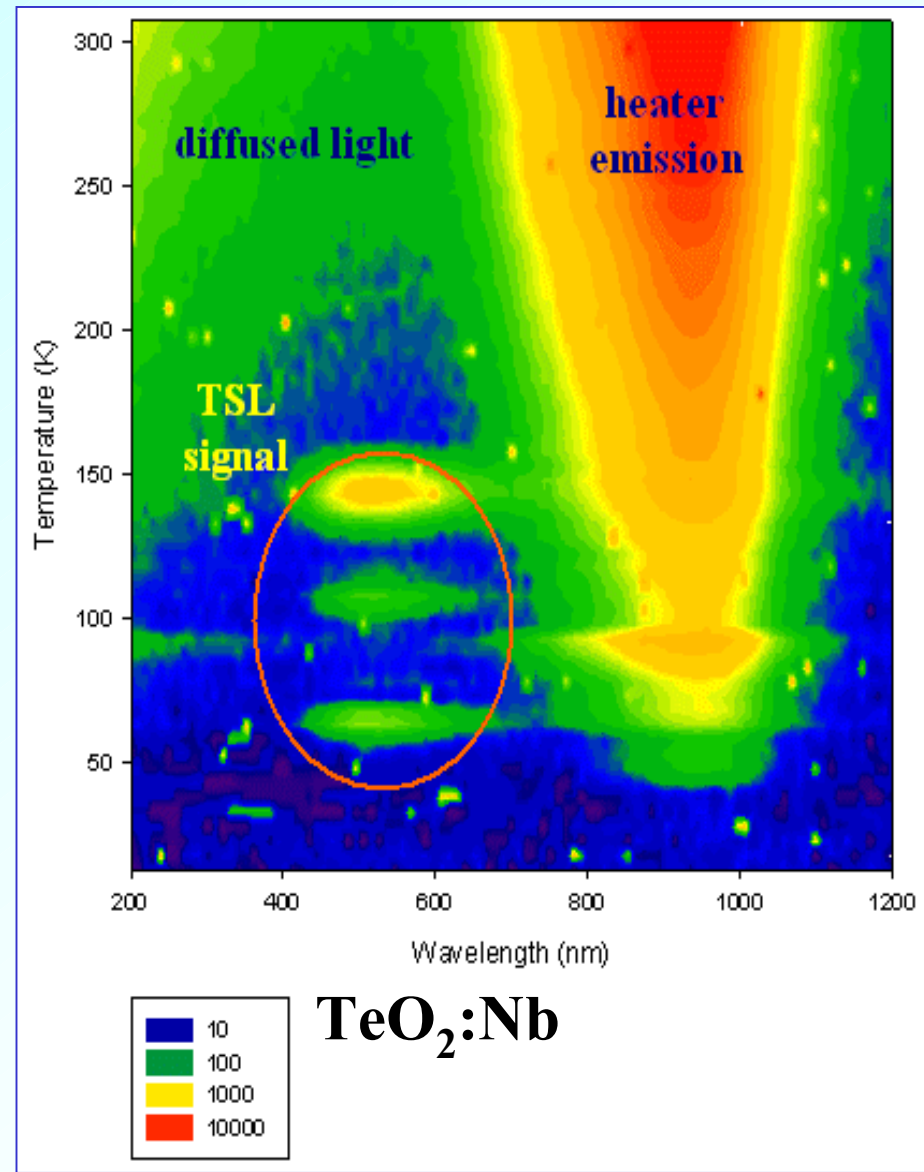
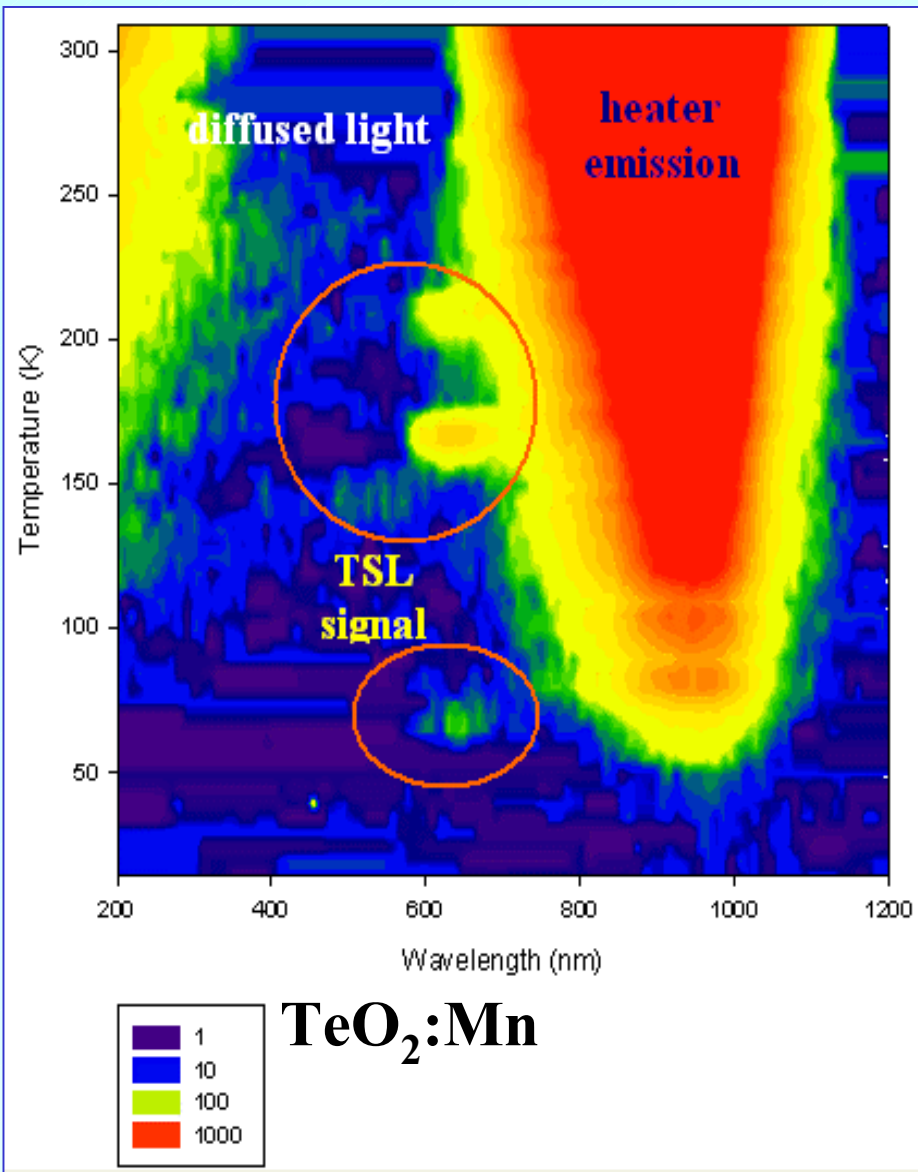
X ray excited steady-state luminescence



X ray excited steady-state luminescence



Thermo-luminescence



after Xray (30 kV) exposure at 10 K

Conclusion and future plans

- *successful growth of doped TeO_2 crystals opens the possibility to realize a hybrid detector (scintillating bolometer) able to separate events due to radioactivity background thus enhancing substantially the sensitivity to neutrinoless DBD measurement.*
- *Mn and Nb doping seems a promising way to enhance the scintillation light yield of TeO_2 crystals.*
- *low temperature photoluminescence measurements are foreseen to find out the nature of scintillation centres*
- *higher dopant concentrations will be tested for a further enhancement of scintillation light yield*