

# Study of Dark Matter with directionality approach using $\text{ZnWO}_4$ crystal scintillators

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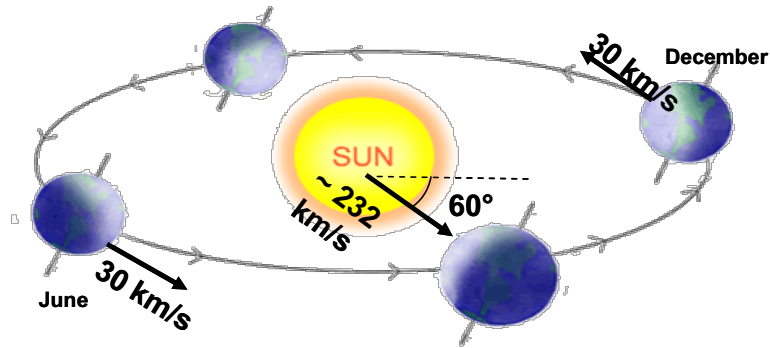
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# Signatures for direct detection experiments

In direct detection experiments to provide a Dark Matter signal identification with respect to the background, a model independent signature is needed.



- Model independent annual modulation: annual variation of the interaction rate due to Earth motion around the Sun which is moving in the Galaxy

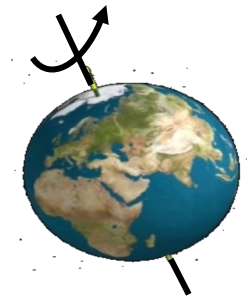
at present the only feasible one, sensitive to many DM candidates and scenarios  
(successfully exploited by DAMA)

- Model independent diurnal modulation: due to the Earth revolution around its axis

2<sup>nd</sup> order effect

- Diurnal variation: daily variation of the interaction rate due to the different Earth depth crossed by the Dark Matter particles

only for high cross sections



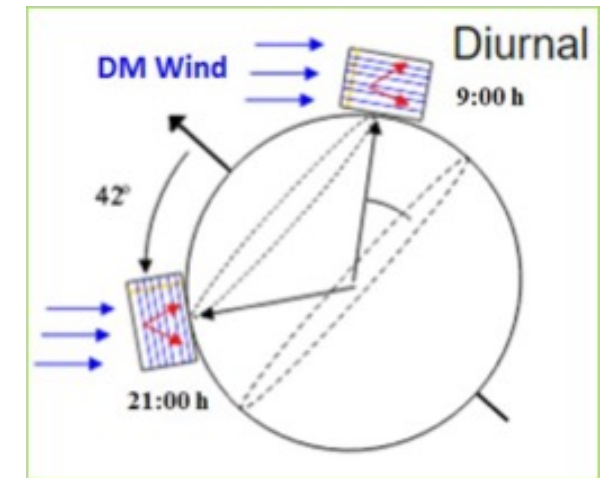
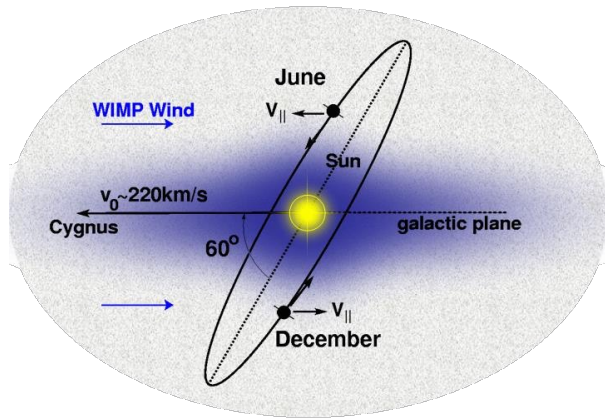
- **Directionality**: correlation of Dark Matter impinging direction with Earth's galactic motion

only for DM candidate particle inducing recoils

# The directionality approach

Based on the study of the correlation between the arrival direction of DM candidates able to induce a nuclear recoil and the Earth motion in the galactic frame.

- Impinging direction of DM particle is (preferentially) opposite to the velocity of the Sun in the Galaxy.
- Due to the Earth's rotation around its axis, the DM particles average direction with respect to an observer on the Earth changes with a period of a sidereal day.
- The direction of the induced nuclear recoil is strongly correlated with that of the impinging DM particle.
- The observation of an anisotropy in the distribution of nuclear recoil direction could give evidence for such DM candidates.



A direction-sensitive detector is needed

# Directionality techniques (R&D stage and ideas)

## Tracking Detectors:

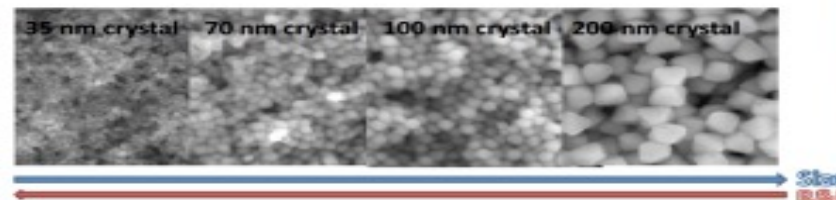
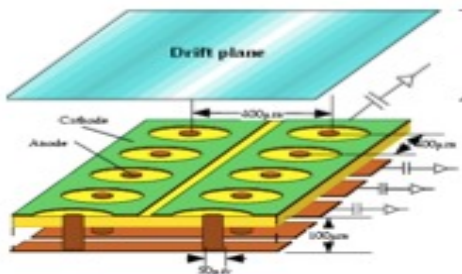
- LP-TPC (DRIFT, NIMAC, DMTPC, NEWAGE, D3, NITEC, CYGNUS, INITIUM)
- Nuclear Emulsions (NEWSdm)
- Ideas: DNA, diamonds

## Detectors using Anisotropic Features:

- Anisotropic crystal scintillators (ADAMO)
- Carbon nanotubes based detectors (PTOLEMY)
- Columnar Recombination in LAr/LXe-TPC (RED)

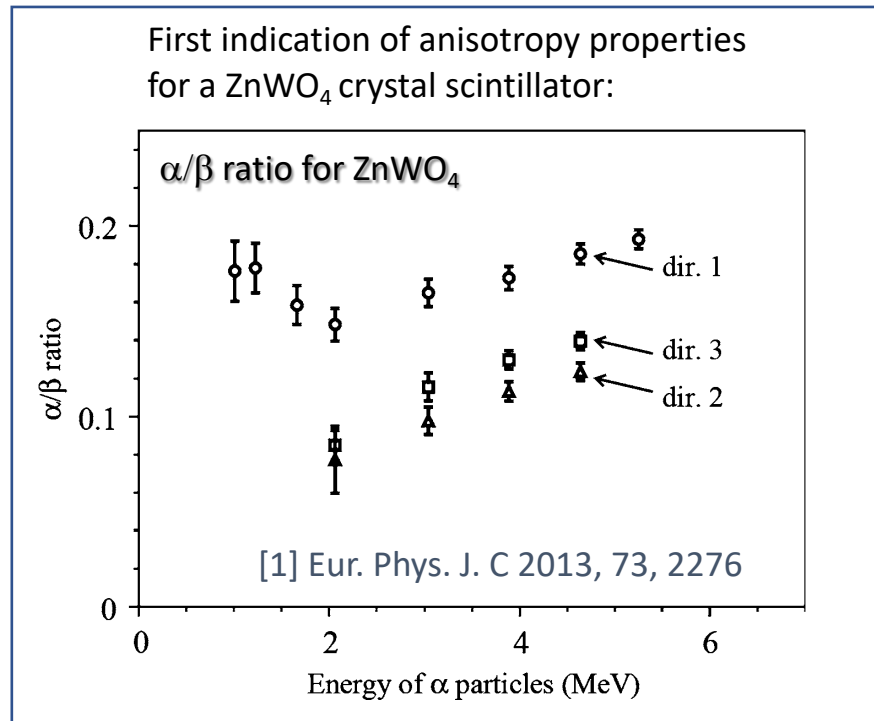
In order to reach a significant sensitivity, a realistic detector experiment needs e.g.:

- **extreme operational stability**
- high radio-purity
- **high mass**
- great spatial resolution (for tracks' detection)
- low energy threshold
- ...



# Anisotropic scintillators: ZnWO<sub>4</sub>

- For **heavy particles** ( $p$ ,  $\alpha$ , nuclear recoils), the light output and the pulse shape depends on the particle impinging direction with respect to the crystal axes;
- For  **$\gamma/e$** , the light output and the pulse shape are isotropic.



## Advantages in the use of ZnWO<sub>4</sub> crystal scintillators:

- Very good anisotropic features;
- High level of radio-purity;
- High light output, that is low energy threshold feasible;
- High stability in the running conditions;
- Sensitivity to small and large mass DM candidate particles;
- Detectors with  $\sim$  kg masses;

Density (g/cm <sup>3</sup> )	7.87
Melting point (°C)	1200
Structural type	Wolframite
Cleavage plane	Marked (010)
Hardness (Mohs)	4–4.5
Wavelength of emission maximum (nm)	480
Refractive index	2.1–2.2
Effective average decay time ( $\mu$ s)	24

# Measurements of $\text{ZnWO}_4$ anisotropic response to nuclear recoils for the ADAMO project

In the framework of the ADAMO project, recent measurements [2] were performed in order to verify the anisotropic response of a  $\text{ZnWO}_4$  crystal scintillator to:

1.  **$\alpha$  particles** : a small  $\text{ZnWO}_4$  crystal ( $10 \times 10 \times 10 \text{ mm}^3$ , with mass of 7.99 g), irradiated by a collimated beam of  $\alpha$  particles from an  $^{241}\text{Am}$  source in the directions along the crystal axes I, II and III.
2. **Oxygen nuclear recoils**: neutron beam of 14 MeV produced by a neutron generator at ENEA-Casaccia.



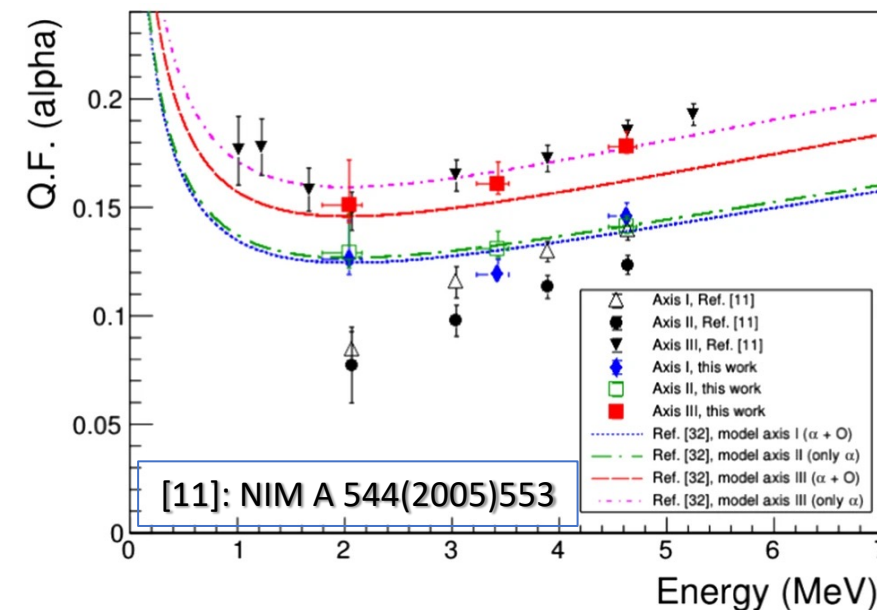
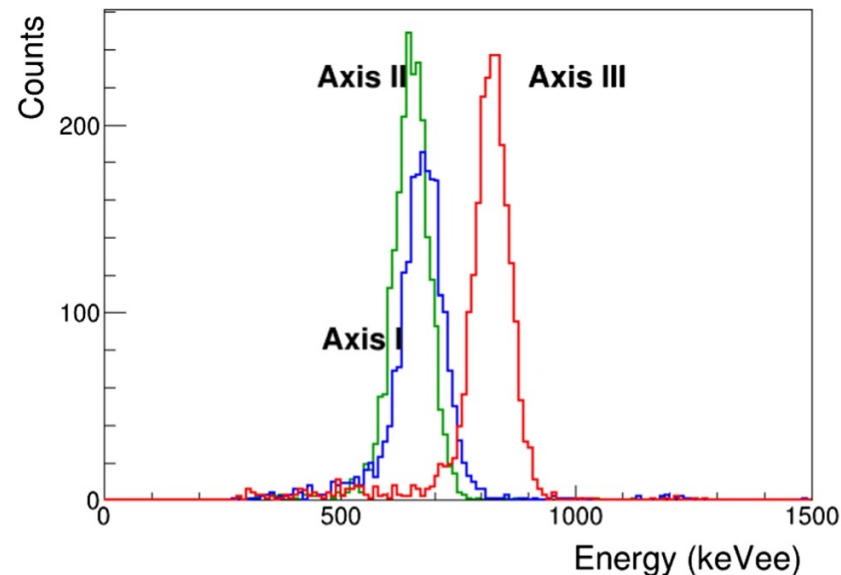
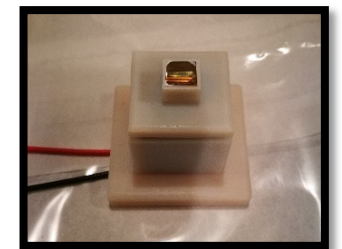
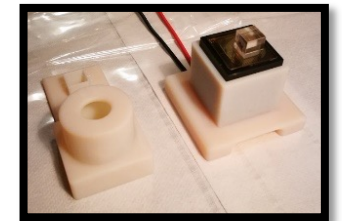
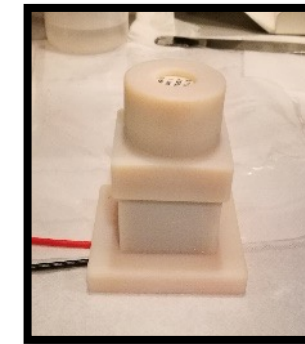
$\text{ZnWO}_4$  crystal =  $10 \times 10 \times 10 \text{ mm}^3$   
(detector of reduced dimensions  
to investigate neutron single-  
scattering)

# Studying the response of the $\text{ZnWO}_4$ with $^{241}\text{Am}$ $\alpha$ source

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Calibration set-up:

- PMT Hamamatsu H11934-200 (transit time  $\approx 5$  ns) +  $\text{ZnWO}_4$
  - LeCroy Oscilloscope 24Xs-A, 2.5 Gs/s, 200MHz bandwidth
  - Pulse profiles acquired in a time window of 100  $\mu\text{s}$
- Crystal irradiated at the same time with  $\gamma$  ( $^{22}\text{Na}$ ) and  $\alpha$  ( $^{241}\text{Am}$ ) sources along the three crystal axes.
- Different  $\alpha$  energies obtained with Mylar foils and measured with Si detector.
- Very efficient PSD capability to discriminate  $\alpha$  and  $\gamma$ .



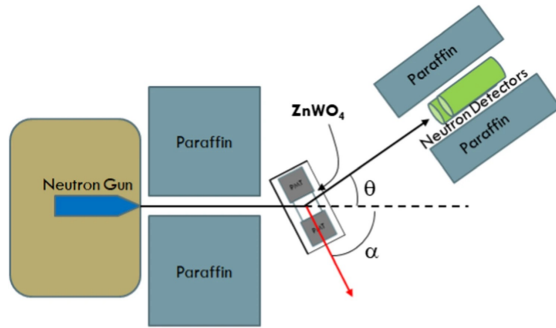
# Studying the response of the $\text{ZnWO}_4$ with a neutron gun

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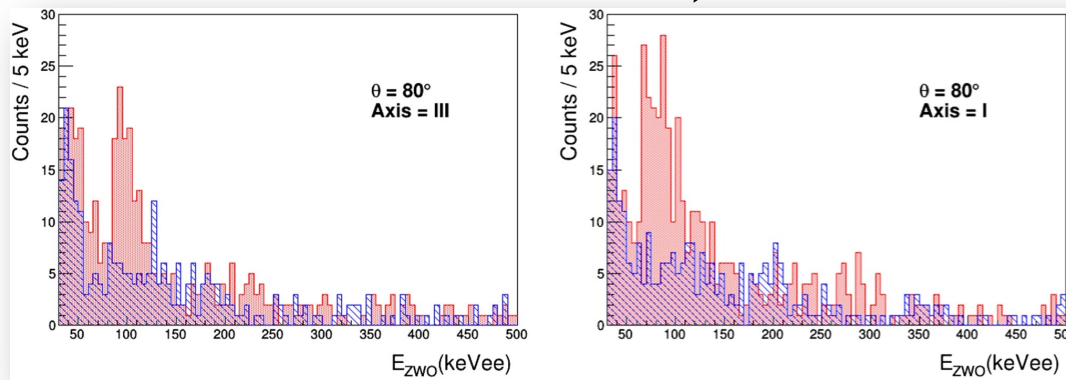
- Set-up:

- ✓  $\text{ZnWO}_4$  Crystal ( $10 \times 10 \times 10 \text{ mm}^3$ )
- ✓ Two Hamamatsu PMTs: HAMA-H11934-200
- ✓ 2 Neutron detectors (Scionix EJ-309)
- ✓ Neutron Gun, Thermo Scientific MP320: 14 MeV neutrons

- Strategy: search for coincidence between a scattered neutron at a fixed angle and scintillation event in  $\text{ZnWO}_4$  occurred in a well defined time window (TOF)
- Once fixed the  $\theta$  angle, the recoil direction and energy are fixed
- Measurements performed at different  $\theta$  angles

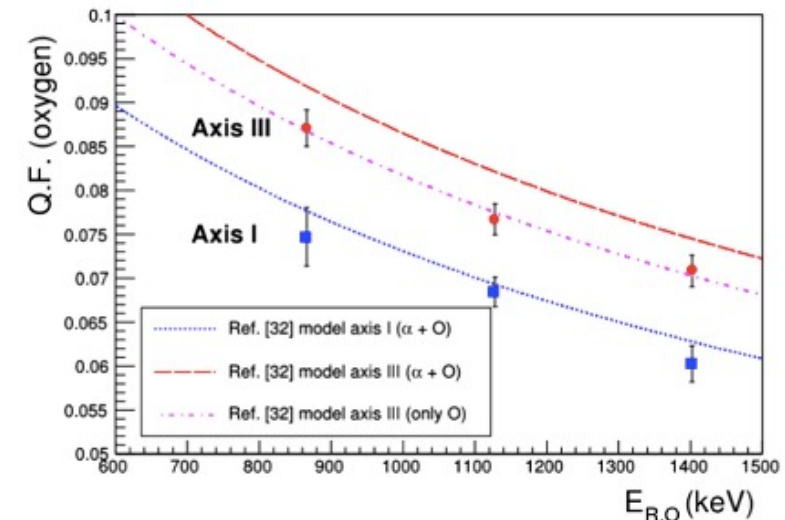


Energy distributions in  $\text{ZnWO}_4$  for coincidence events when neutrons are identified in EJ-309 and two TOF windows are considered ( $\theta=80^\circ$ ) to consider the **neutron induced recoils** and to characterize the **random coincidences**



The anisotropy is significantly evident also for oxygen nuclear recoils in the energy region down to hundreds keV at **5.4  $\sigma$  confidence level**.

## First evidence at low energy





# Optical and scintillation properties of advanced ZnWO<sub>4</sub> crystal scintillators

High optical and scintillation properties ZnWO<sub>4</sub> crystal scintillators were developed by using the **low-thermal gradient Czochralski technique** after an extended R&D that included variation of the compound stoichiometry, **using** of initial WO<sub>3</sub> of **different producers** and **additionally purified**, utilization of **single** and **double crystallization with and without annealing** of the grown boules.

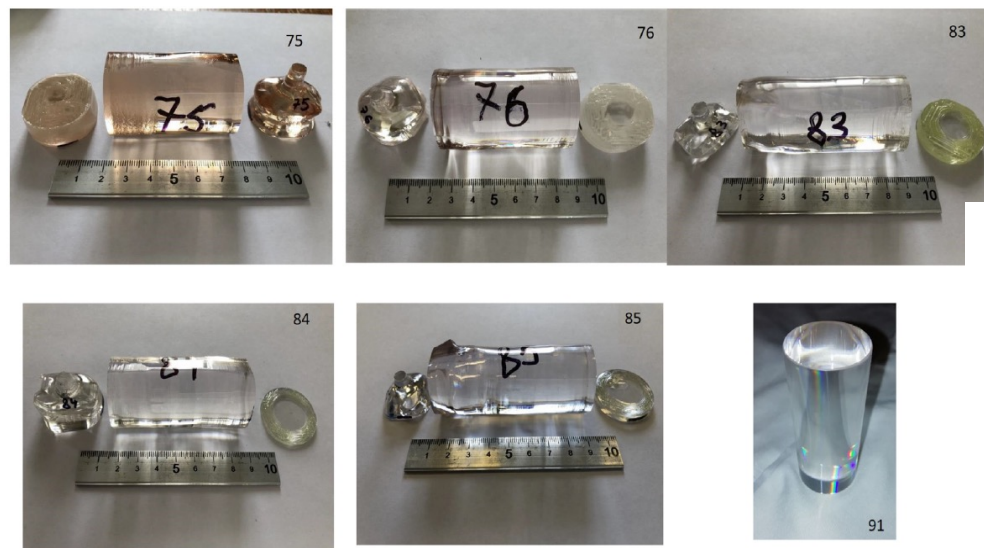
NIMA1029(2022)166400

**Table 1**  
The samples of ZnWO<sub>4</sub> crystals used in the present study and the boules of origin.

Crystal boule	Sample size (mm <sup>3</sup> )	Number of crystallizations	WO <sub>3</sub> tungsten trioxide	Compound stoichiometry
No. 75	10 × 10 × 2 ∅30 × 60	Double	NIIC II	+0.3% of WO <sub>3</sub>
No. 76	10 × 10 × 2 ∅30 × 60	Double	Nippon Tungsten Co., Ltd	+0.25% of ZnO
No. 83	10 × 10 × 2 ∅30 × 60	Single, annealed	NIIC I	+0.15% of WO <sub>3</sub>
No. 84	10 × 10 × 2 ∅30 × 60	Single, annealed	NIIC I	Stoichiometric
No. 85	10 × 10 × 2 ∅30 × 60	Single, annealed	Japan New Metals Co., Ltd	Stoichiometric
No. 91	∅30 × 67	Single, annealed	NIIC I	Stoichiometric
No. 94	∅30 × 31 ∅30 × 32	Single, annealed	NIIC I	Stoichiometric

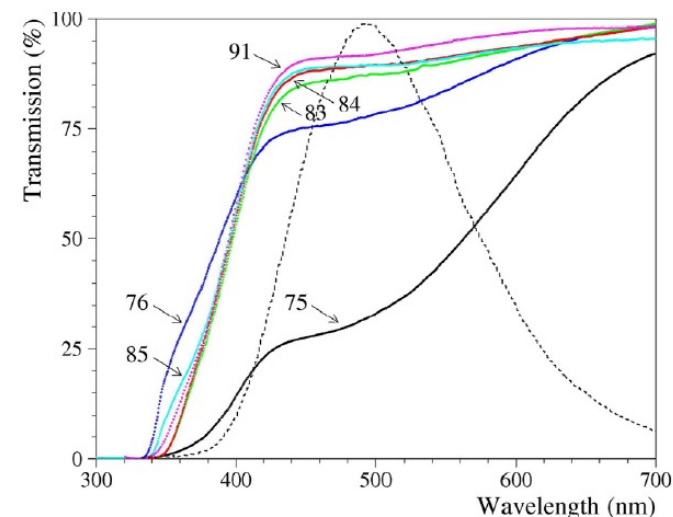
Several samples of tungsten oxide of different origin were utilized:

- **NIIC I**: synthesized at the NIIC (Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia) with Si concentration <50 ppm and concentration of transition metals less than 1 ppm
- **NIIC II**: purified at the NIIC using an additional process of sublimation of tungsten chlorides;
- manufactured by **Nippon Tungsten Co., Ltd.** (Japan);
- manufactured by **Japan New Metals Co., Ltd** with a maximum Fe content <1 ppm and a maximum Mo content less than 10 ppm.



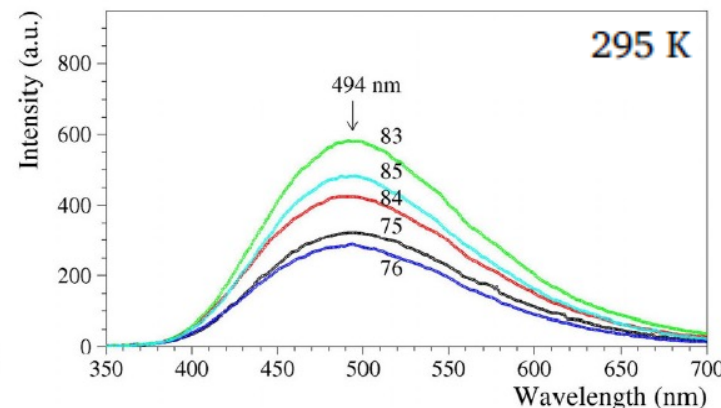
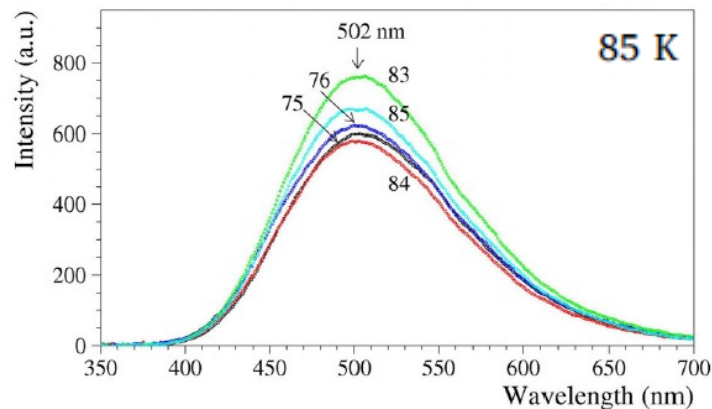
The **transmission spectra agree** with the **literature** data. However, the transmission varies substantially **depending** on the sample **production protocol**. In particular, the samples produced by double crystallization (samples 75, 76) are definitely of worse optical quality

## Optical transmission



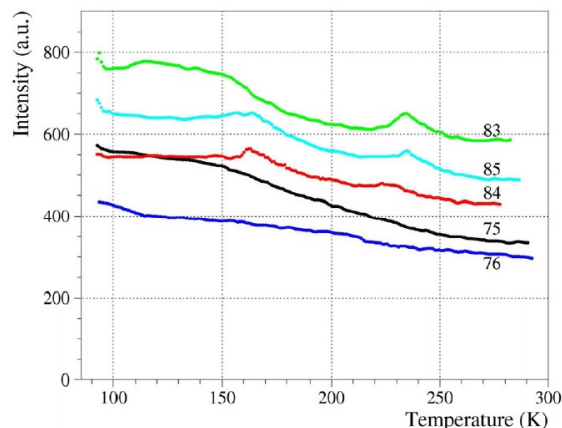
## Luminescence under X-ray excitation and scintillation properties

Emission spectra under X-ray irradiation at 85 K and 295 K

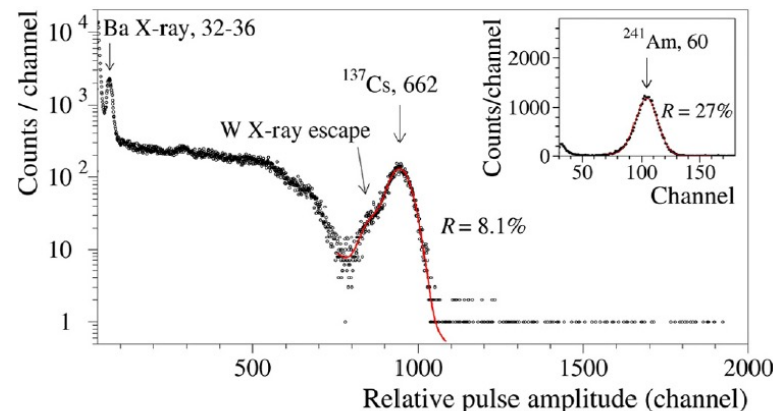


### Bigger spread at 295 K

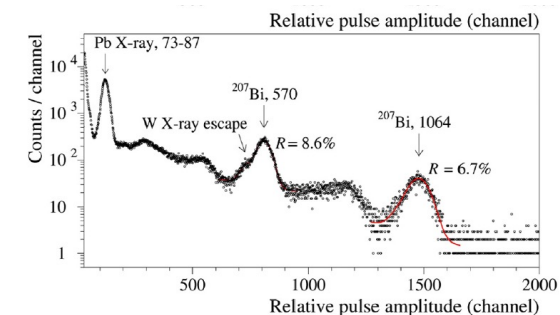
The difference can be explained by presence of non-radiative recombination centers (caused by defects of different nature) that compete with the radiative recombination centers.



The luminescence intensity increased with temperature decrease, in agreement with the data of other studies

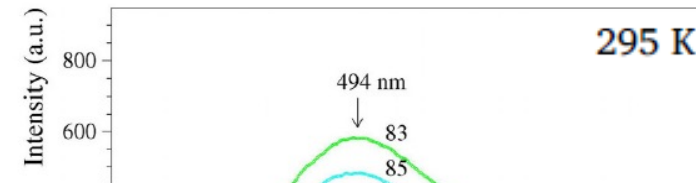
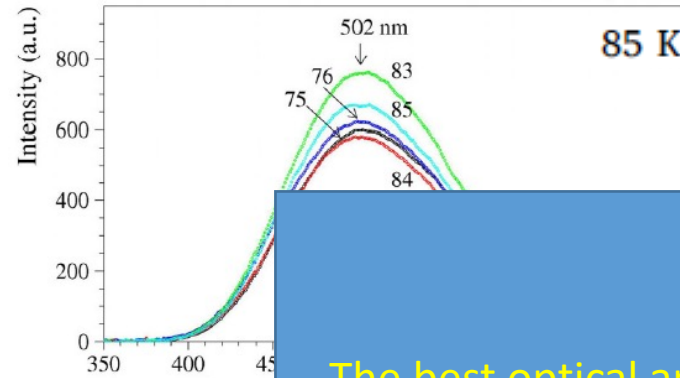


The energy spectra of  $\gamma$ -ray quanta of <sup>137</sup>Cs and <sup>207</sup>Bi measured by scintillation detector with the ZnWO<sub>4</sub> crystal sample No. 84. Energy spectrum of  $\gamma$ -ray quanta of <sup>241</sup>Am is shown in Inset. Energy of X-ray and  $\gamma$ -ray quanta is in keV.



## Luminescence under X-ray excitation and scintillation properties

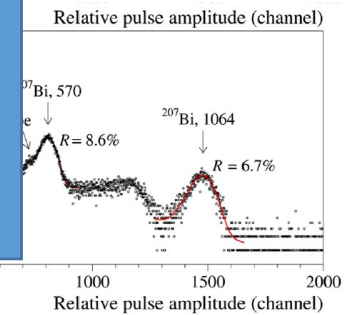
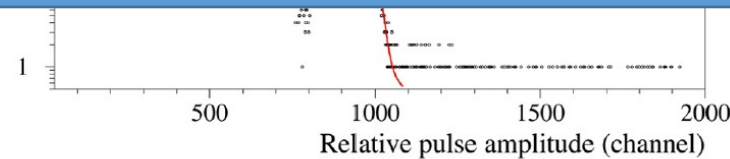
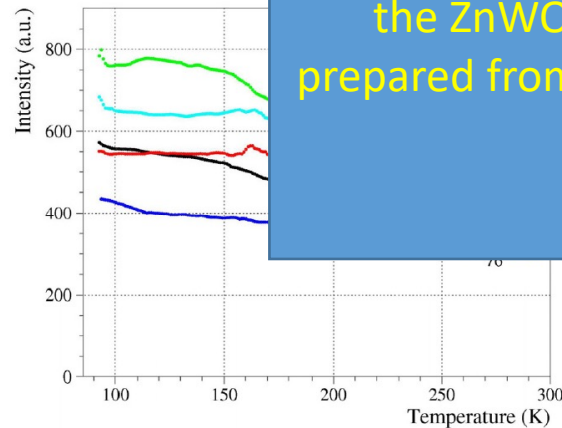
Emission spectra under X-ray irradiation at 85 K and 295 K



**Bigger spread at 295 K**

The difference can be explained by presence of non-radiative recombination centers (caused of different nature) compete with the radiative recombination centers.

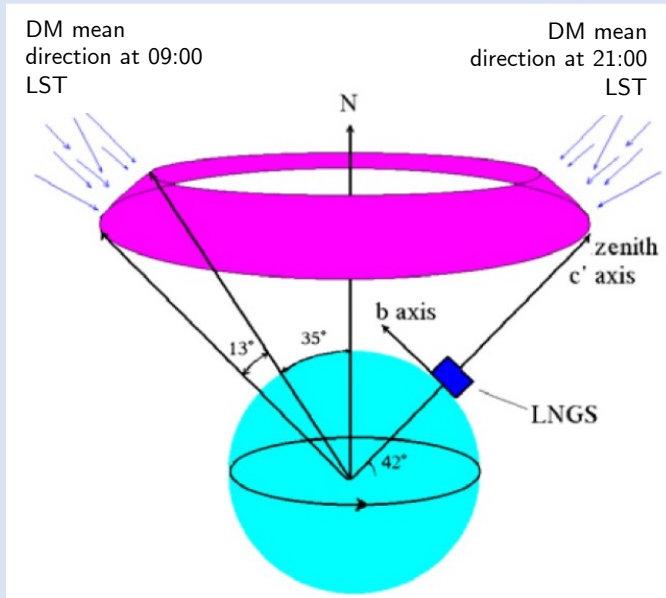
The best optical and scintillation characteristics were obtained with ZnWO<sub>4</sub> crystal samples grown by **single crystallization** from the ZnWO<sub>4</sub> compound of the **stoichiometric composition** prepared from deeply purified WO<sub>3</sub>, **annealed** in air atmosphere.



The luminescence intensity increased with temperature decrease, in agreement with the data of other studies

The energy spectra of  $\gamma$ -ray quanta of <sup>137</sup>Cs and <sup>207</sup>Bi measured by scintillation detector with the ZnWO<sub>4</sub> crystal sample No. 84. Energy spectrum of  $\gamma$ -ray quanta of <sup>241</sup>Am is shown in Inset. Energy of X-ray and  $\gamma$ -ray quanta is in keV.

# Example of expected signal

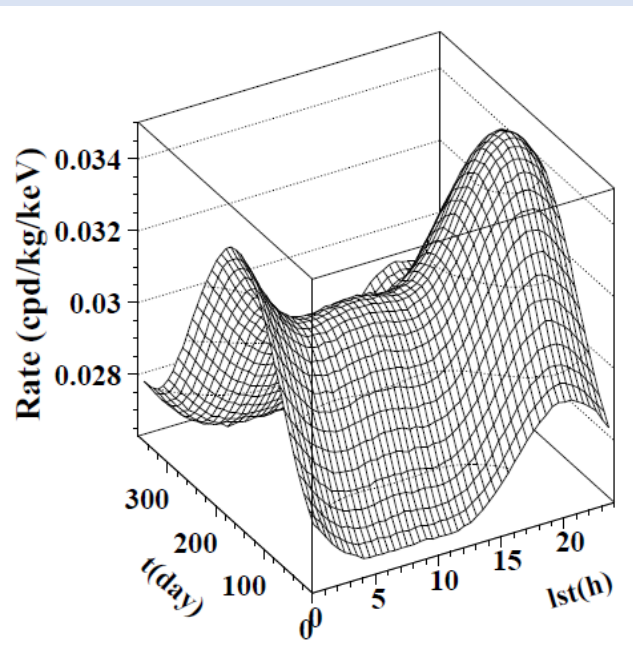


- It is very convenient to consider an experiment performed at the LNGS latitude (**42°27'N**):
- ❖ DM particles come mainly from the top at 21.00 h LST
- ❖ 12 h later they come from the North and parallel to the horizontal plane

- If we arrange the  $\text{ZnWO}_4$  crystal axis so that:
    - The one with the largest light output is vertical and
    - the one with the smallest light output points north
- ⇒ We obtain the maximum range of variability of the anisotropic detector response during a sidereal day

Absolute maximum rate is at day 152 and at 21h LST (when the DM flux is at maximum and the DM preferential arrival direction is near the zenith)

Expected rate as a function of sidereal time and days of the year



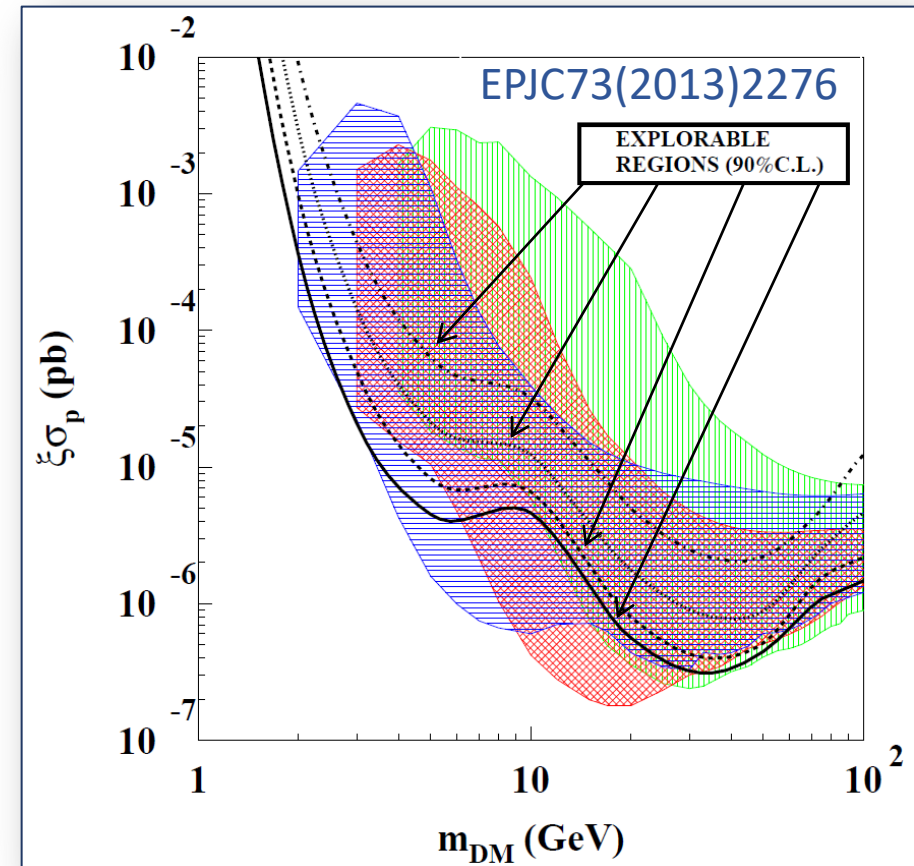
[2-3] keV  
 $\sigma_p = 5 \times 10^{-5}$  pb  
 $m_{\text{DM}} = 10$  GeV

# ADAMO project: example of reachable sensitivity in a given scenario

Assumptions:

- simplified model framework
- 200 kg of  $\text{ZnWO}_4$
- 5 years of data taking
- 2 keVee threshold
- four possible time independent background levels in the low energy region:

- $10^{-4}$  cpd/kg/keV —————
- $10^{-3}$  cpd/kg/keV - - - - -
- $10^{-2}$  cpd/kg/keV .....  
.....
- 0.1 cpd/kg/keV — · — · — · —

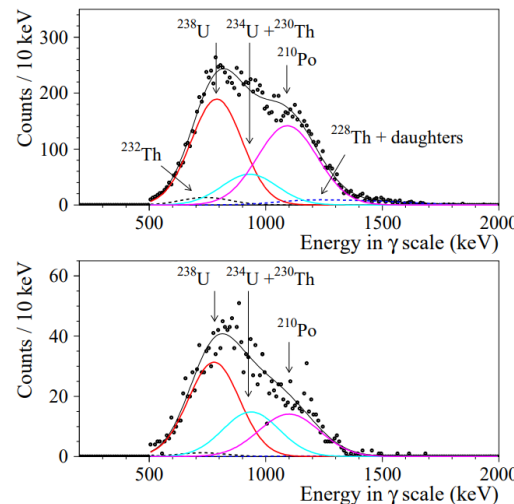


The directionality approach can reach in the given scenario a sensitivity to the cross section at level of  $10^{-5} - 10^{-7}$  pb, depending on the particle mass.

Allowed regions (green, red, blue) obtained with a corollary analysis of the  $9.3\sigma$  C.L. DAMA model independent result in terms of scenarios for the DM candidates considered here.

# ZnWO<sub>4</sub> – work in progress...

- ❖ A cryostat for low temperature measurement with scintillation detectors has been realized
- ❖ Test of the cryostat is in progress
- ❖ Lowering the energy threshold (new PMT with higher QE optimized to the fluorescence light emission and temperature operation)



An example of the radio-purity improvements after the recrystallization

- ❖ New measurements of anisotropy at low energy with MP320 Neutron Generator ( $E_n = 14$  MeV) at ENEA-Casaccia is ongoing
- ❖ Further improvement of the radio-purity

# Conclusions

- Directionality Dark Matter experiments could obtain, with a completely different new approach, further evidence for the presence of DM candidates inducing nuclear recoils in the galactic halo and/or provide complementary information on the nature and interaction type of DM particle candidates.
- Several TPC-based detectors are in the R&D stage. Other potential ideas have shortly been listed.
- The anisotropic  $\text{ZnWO}_4$  detectors are promising to investigate the directionality for DM candidates inducing nuclear recoils.
- First evidence of anisotropy in the response of  $\text{ZnWO}_4$  crystal scintillator to low energy nuclear recoils reported.
- The data presented here confirm the anisotropic response of the  $\text{ZnWO}_4$  crystal scintillator to  $\alpha$  particles in the MeV energy region. The anisotropy is significantly evident also for oxygen nuclear recoils in the energy region down to some hundreds keV at  $5.4 \sigma$  confidence level.