

# ZnWO<sub>4</sub> anisotropic scintillators for directionality technique



**CYGNUS 2019 workshop on directional DM  
detection**

**July 10-12 2019, Rome (Italy)**

**Dr. A. Di Marco**  
**INFN Roma “Tor Vergata”**



# Introduction

- Development of low-background anisotropic detectors is of great interest in many applicative fields

In particular, it offers an unique way to further investigate those Dark Matter (DM) candidate particles able to induce just nuclear recoils, through the so-called directionality approach

- In this talk we summarize the relevance of a low background pioneer experiment (Anisotropic detectors for DArk Matter Observation, ADAMO) to effectively exploit deep underground the directionality approach by using highly radio-pure anisotropic  $\text{ZnWO}_4$  scintillators

# Velocity of a detector in a terrestrial laboratory

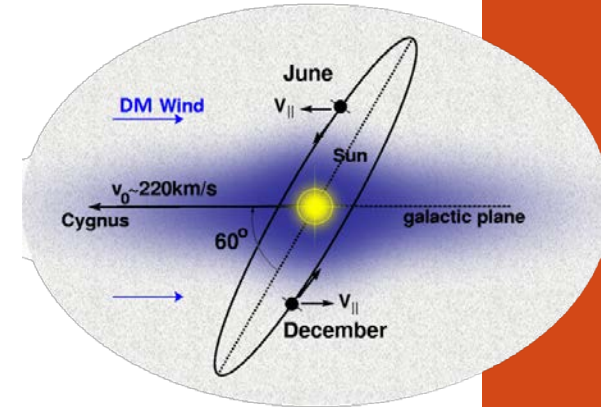
$$\vec{v}_{lab}(t) = \vec{v}_{LSR} + \vec{v}_{\odot} + \vec{v}_{rev}(t) + \vec{v}_{rot}(t)$$

$\vec{v}_{LSR}$  Velocity of the Local Standard of Rest (LSR) due to Galaxy rotation

$\vec{v}_{\odot}$  Sun peculiar velocity with respect to LSR

$\vec{v}_{rev}(t)$  Velocity of the revolution of the Earth around the Sun

$\vec{v}_{rot}(t)$  Velocity of the rotation of the Earth around its axis @ lab (lat, lng)



The **Sun velocity**,  $\vec{v}_S$ , in the *Galactic Coordinate system* is:

$$\vec{v}_{LSR} = (0, v_0, 0) \quad (v_0 = 220 \pm 50 \text{ km/s})$$

$$\vec{v}_{\odot} = (9, 12, 7) \text{ km/s}$$

$$\Rightarrow \vec{v}_S = \vec{v}_{LSR} + \vec{v}_{\odot} = (9, 232, 7) \text{ km/s}$$

The **Earth revolution velocity** in the *Ecliptic plane* ( $\hat{e}_1^{ecl}, \hat{e}_2^{ecl}$ ) is:

$$\vec{v}_{rev}(t) = V_{Earth} (\hat{e}_1^{ecl} \sin \lambda(t) - \hat{e}_2^{ecl} \cos \lambda(t))$$

$$\lambda(t) = \omega(t - t_{equinox}); \quad \omega = \frac{2\pi}{T}; \quad T=1y$$

$$(V_{Earth} \approx 29.8 \text{ km/s}; \quad t_{equinox} \approx \text{March 21})$$

The **Earth rotation velocity** in the *Equatorial plane* ( $\hat{e}_1^{ecs}, \hat{e}_2^{ecs}$ ) is:

$$\vec{v}_{rot}(t) = -V_r (\hat{e}_1^{ecs} \sin \delta(t) - \hat{e}_2^{ecs} \cos \delta(t))$$

$$\delta(t) = \omega_{rot} t; \quad \omega_{rot} = \frac{2\pi}{T_d}; \quad T_d = 1 \text{ sidereal day}$$

(Here t is the local sidereal time, LST)

In *Galactic Coordinate System*:

$$\left\{ \begin{array}{l} \hat{e}_1^{ecl} = (-0.05487, 0.49411, -0.86767) \\ \hat{e}_2^{ecl} = (-0.99382, -0.11100, -0.00035) \\ \hat{e}_3^{ecl} = (-0.09648, 0.86228, 0.49715) \end{array} \right.$$

$$\hat{e}_3^{ecl} \cdot (0, 0, 1) = 0.49715.$$

$\Rightarrow 60^\circ$  inclination w.r.t. galactic plane

$$\left\{ \begin{array}{l} \hat{e}_1^{ecs} = (-0.05487, 0.49411, -0.86767) \\ \hat{e}_2^{ecs} = (-0.87344, -0.44483, -0.19808) \\ \hat{e}_3^{ecs} = (-0.48384, 0.74698, 0.45599) \end{array} \right.$$

@ LNGS ( $\phi_0 = 42^\circ 27' N$ ;  $\lambda_0 = 13^\circ 34' E$ )

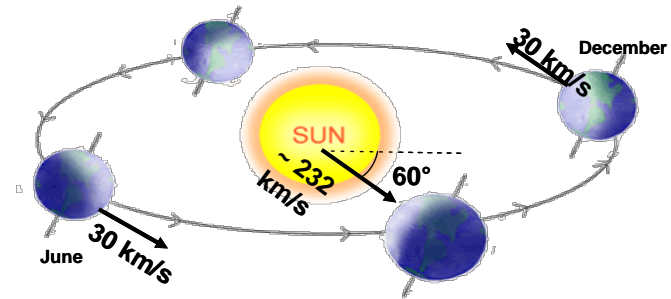
$$V_r = V_{eq} \cos \phi_0 = 0.3435 \text{ km/s}$$

$$(V_{eq} = 0.4655 \text{ km/s})$$

# Model independent signatures for DM direct detection

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)



$$\hat{v}_S \cdot \vec{v}_{rev}(t) = V_{Earth} A_m \cos[\omega(t - t_0)]$$

$$A_m \approx 0.489;$$

$$t_0 = t_{equinox} + 73.25 \text{ solar days}$$

$$([71.8, 74.2] \text{ d when varying } v_0 \text{ in } [170, 270] \text{ km/s})$$



- Model independent Diurnal modulation: due to the Earth revolution around its axis (2<sup>nd</sup> order effect)

$$\hat{v}_S \cdot \vec{v}_{rot}(t) = V_r A_d \cos[\omega_{rot}(t - t_d)]$$

$$A_d \approx 0.671;$$

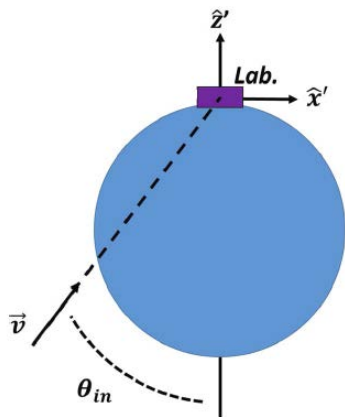
$$t_d = 14.92 \text{ h LST}$$

$$([14.84, 14.97] \text{ h when varying } v_0 \text{ in } [170, 270] \text{ km/s})$$

⇒ The ratio,  $R_{dy}$ , of the DM diurnal modulation amplitude ( $S_d$ ) over the DM annual modulation amplitude ( $S_m$ ) is a model independent constant:

$$R_{dy} = \frac{S_d}{S_m} = \frac{V_r A_d}{V_{Earth} A_m} \approx 0.016 \text{ at LNGS latitude}$$

# Assuming DM candidates inducing just nuclear recoils, useful approaches



- **Shadow Effect:** daily variation of the interaction rate due to different Earth depth crossed by such DM candidates (**only for high  $\sigma$** )

The thickness crossed before reaching a laboratory depends on the particle impinging angle  $\theta_{in}$

$$\langle \theta_{in} \rangle = \pi - \langle \theta \rangle$$

@LNGS The Earth shielding is Max at  $\sim 9:00$  h LST and Min at  $\sim 21:00$  h LST

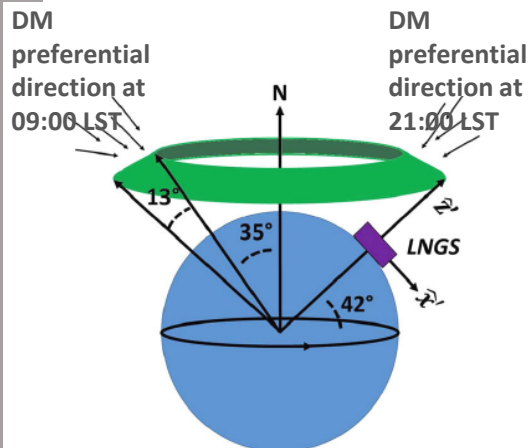
- **Directionality**

Based on diurnal variation of apparent DM wind arrival direction

Study of the correlation between the arrival direction of Dark Matter candidates inducing nuclear recoils and the Earth motion in the galactic frame

Direction of the induced nuclear recoil is strongly correlated with that of the considered impinging DM candidate

The observation of an anisotropy in the distribution of nuclear recoil direction could give evidence for such candidates



**direction-sensitive detector**

# All the diurnal effects are based on sidereal time

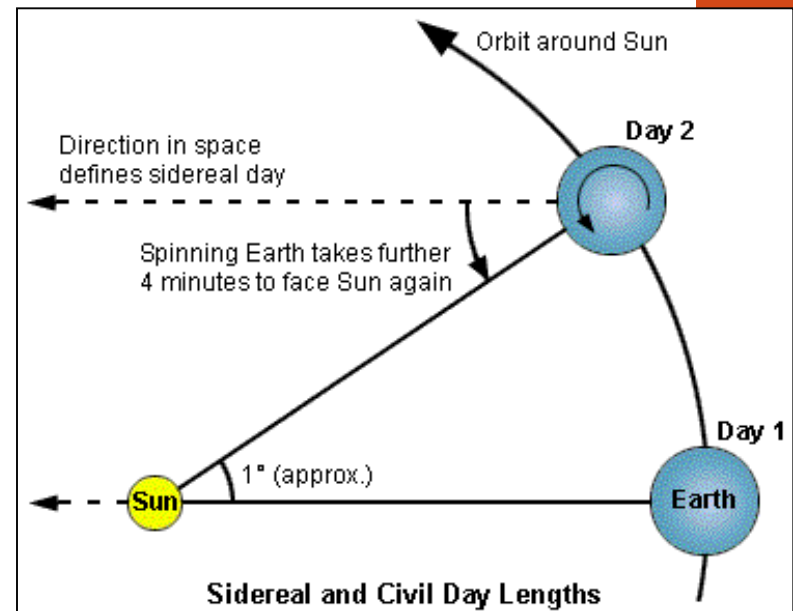
- Diurnal modulation effect due to Earth rotation
- Shadow effect
- Directionality

All effects with different phases but the same period:

$$T = 1 \text{ sidereal day}$$

$$1 \text{ solar day} \cong 1.00274 \text{ sidereal days}$$
$$(365.25 \text{ solar days} \cong 366.25 \text{ sidereal days})$$

**Local sidereal time is 00:00 when the vernal equinox crosses the local meridian**



⇒ Side effects related to **solar time**, even if with a similar phase, could not have any role in the interpretation of a possible signal

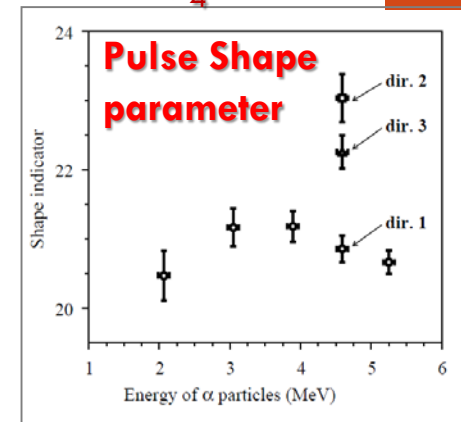
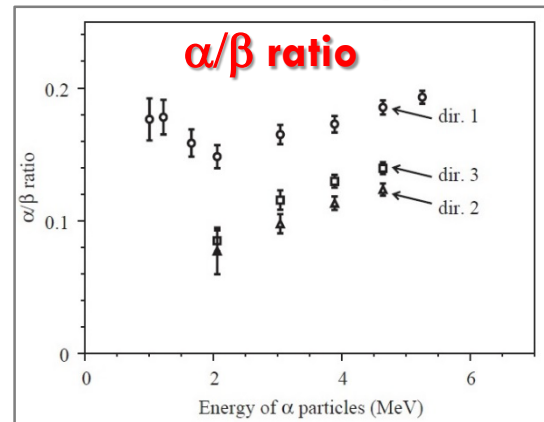
# Directionality sensitive detectors overcoming the track measurement difficulties: anisotropic scintillators

- Firstly proposed in [Il Nuovo Cim. C 15 (1992) 475], revisited in [EPJC28(2003)203] and then in [EPJC73(2013)2276]

- Anisotropic Scintillator:

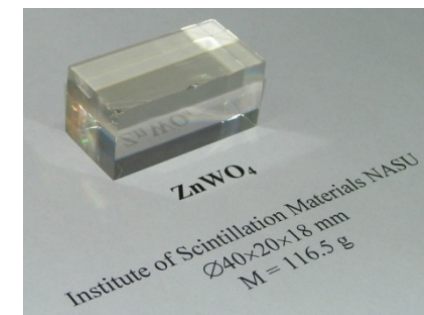
- **for heavy particles** the light output and the pulse shape depends on the particle impinging direction with respect to the crystal axes
- **for  $\gamma/e$**  the light output and the pulse shape are instead isotropic

## $\alpha$ Particles in $ZnWO_4$



The variation of the response of an anisotropic scintillator during sidereal day can allow to point out the presence of a DM signal due to candidate inducing just nuclear recoils overcoming the problems due to the shortness of the recoil tracks

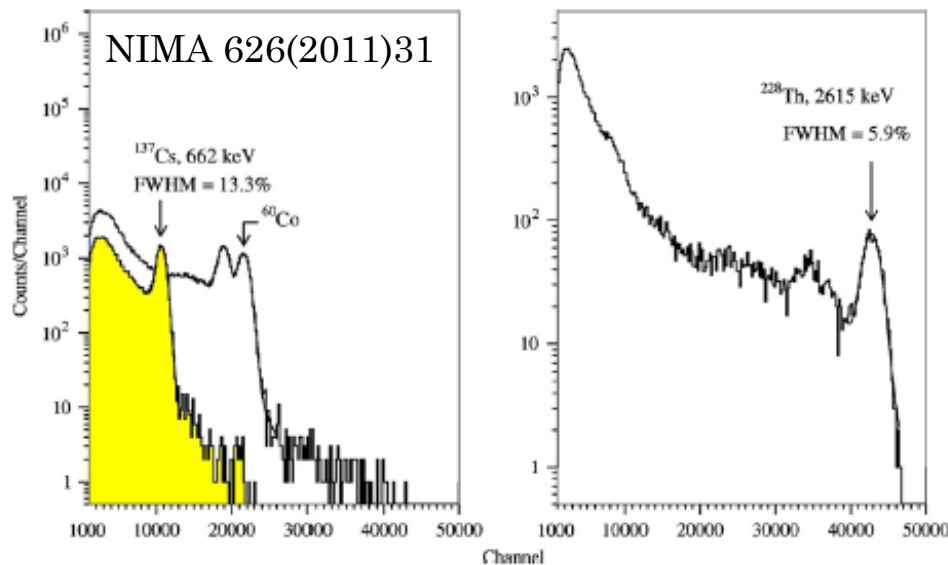
- **$ZnWO_4$  anisotropic scintillator:** a very promising detector (Eur. Phys. J. C 73 (2013) 2276)



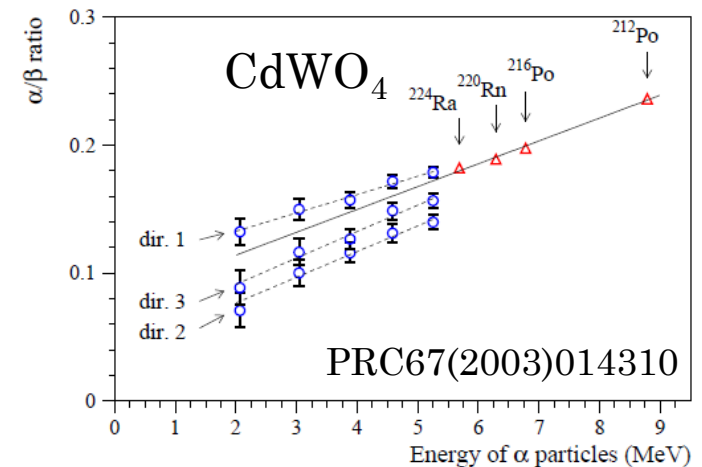
# Anisotropic scintillators

In some scintillation crystals with anisotropic structure,  $\alpha/\beta$  ratio depends on the direction of  $\alpha$  particles relatively to crystal axes:

- anthracene [P. Schuster, IEEE Trans. Nucl. Sci. 63, 1942 (2016)]
- stilbene [P. Schuster, E. Brubaker, Nucl. Instr. Meth. A 859, 95 (2017)]
- $\text{CdWO}_4$  [F.A. Danevich et al., Phys. Rev. C67, 014310(2003)]
- $\text{ZnWO}_4$  [F.A. Danevich et al., Nucl. Instr. Meth. A544, 553 (2005)]
- $\text{MgWO}_4$  [F.A. Danevich et al., Nucl. Instr. Meth. A608, 107(2009)]



$\text{ZnWO}_4$  calibrations



For further theoretical details see e.g. J.B. Birks, "The Theory and Practice of Scintillation Counting" (1964). London: Pergamon Press



# Advantages of the $\text{ZnWO}_4$ crystal

- ✓ 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



Eur. Phys. J. C 73 (2013) 2276

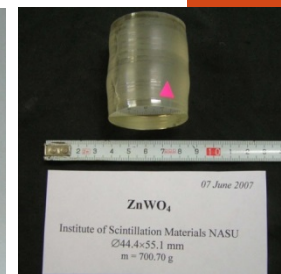
<i>Density (g/cm<sup>3</sup>)</i>	7.87
<i>Melting point (°C)</i>	1200
<i>Structural type</i>	Wolframite
<i>Cleavage plane</i>	Marked (010)
<i>Hardness (Mohs)</i>	4–4.5
<i>Wavelength of emission maximum (nm)</i>	480
<i>Refractive index</i>	2.1–2.2
<i>Effective average decay time (μs)</i>	24

# ZnWO<sub>4</sub> crystal scintillators in DAMA-INR Kiyv collaboration

- Low background ZnWO<sub>4</sub> crystal scintillators with large volume and good scintillation properties realized
- Various detectors with mass **0.1-0.7 kg** realized by exploiting different materials and techniques
- For example in [PLB658(2008)193] detector installed in a cavity (filled up with high-pure silicon oil) in the central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The light-guides were faced by 2 low-background PMTs

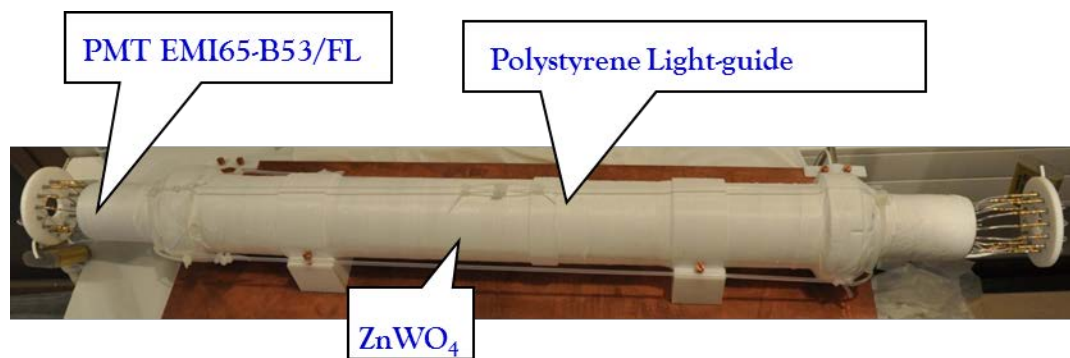
PLB658(2008)193,  
NPA826(2009)256,  
NIMA626-627(2011)31,  
JP38(2011)115107

Crystal scintillator	Size (mm)	Mass (g)
ZWO-1	20 × 19 × 40	117
ZWO-2	∅44 × 55	699
ZWO-2a	∅44 × 14	168

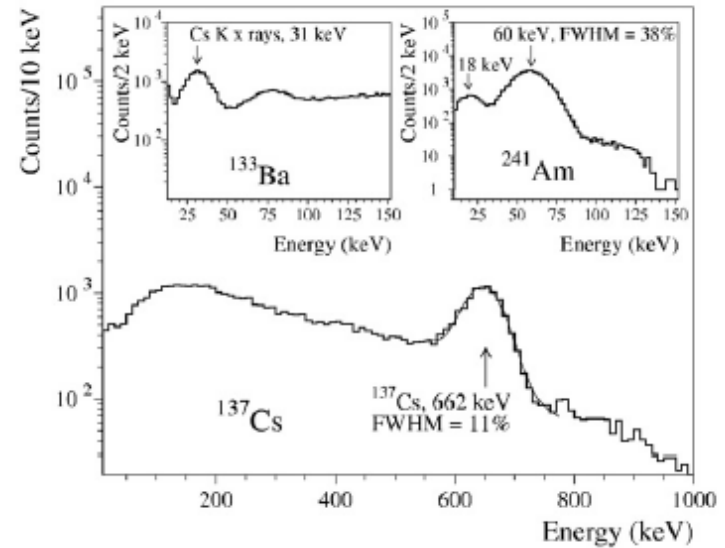
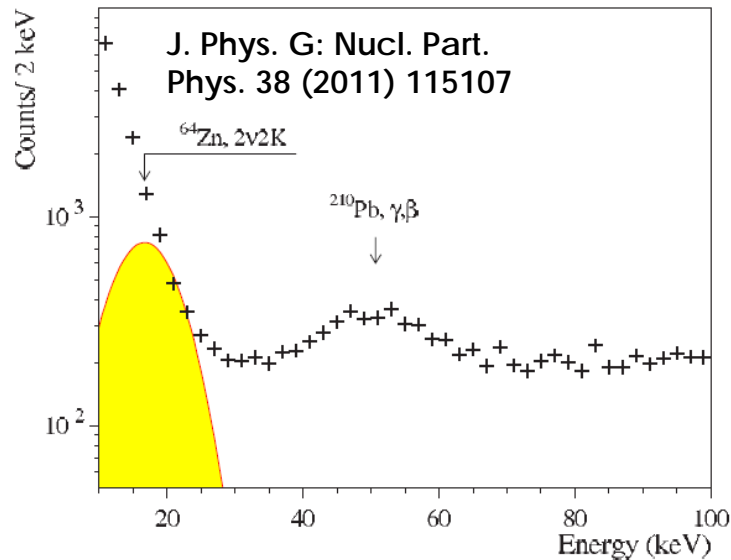


Main aim of the measurements:

- study of the properties of ZnWO<sub>4</sub>
- search for 2β processes in Zinc and Tungsten isotopes ( $T_{1/2} \sim 10^{18} - 10^{21}$  yr)



Reached energy threshold 10 keV (experiment not optimized for the low energy region)



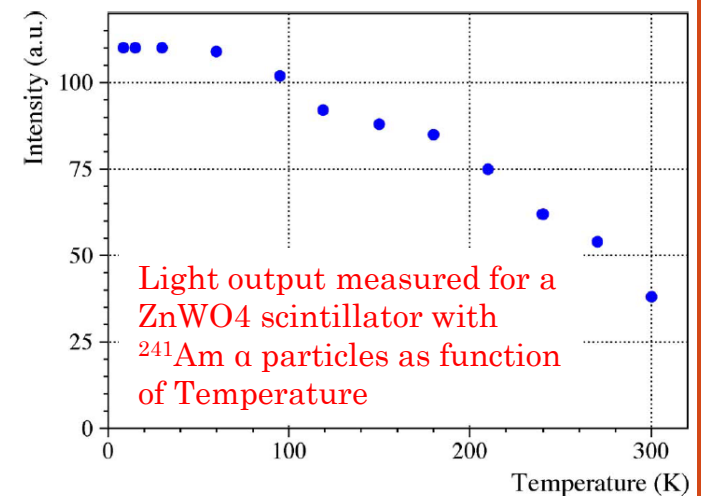
FWHM (8.8–14.6)% @662 keV

Improvements of the energy threshold by:

- ✓ coupling 2 PMTs in coincidence at single p.h.e. level
- ✓ decreasing operational temperature
- ✓ crystal in silicone oil (light collection improvement ~40%)
- ✓ using silicon photodiodes, APD, SiPM, etc. or with a combination of the previous points

**Lower threshold feasible**

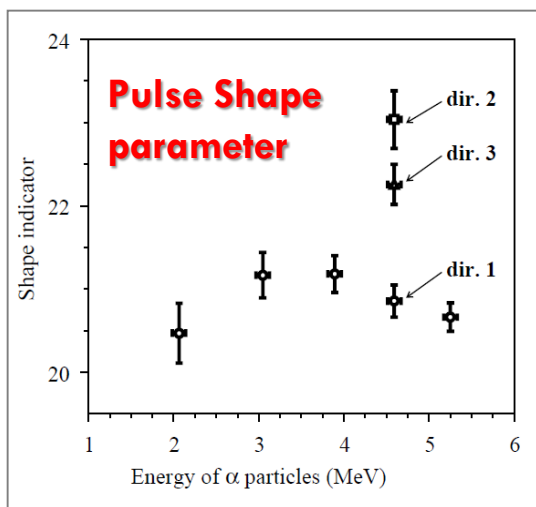
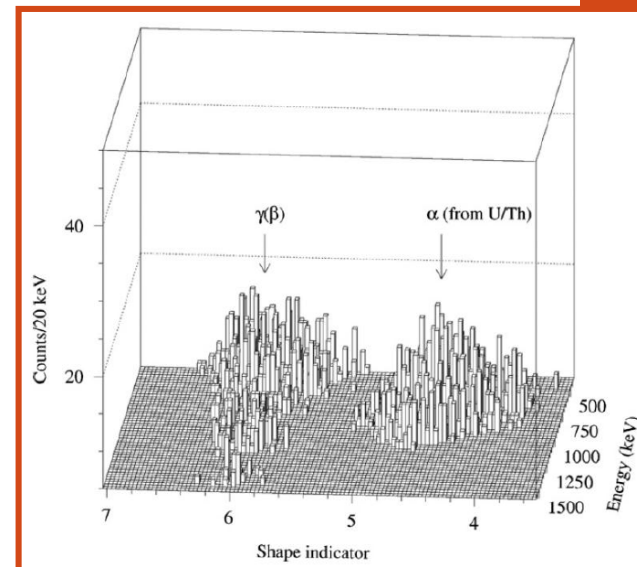
IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 56, NO. 3, JUNE 2009



# PSD capability of the $\text{ZnWO}_4$ crystal scintillator

The dependence of the pulse shapes on the type of irradiation in the  $\text{ZnWO}_4$  scintillator allows one to discriminate  $\beta(\gamma)$  events from those induced by  $\alpha$  particles and to identify the  $\alpha$  background

Pulse Shape analysis: possible to point out the anisotropic behavior for low energy nuclear recoils

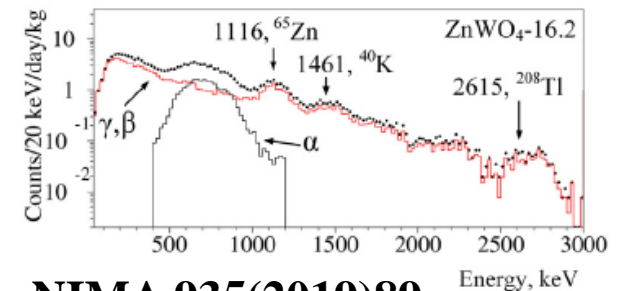
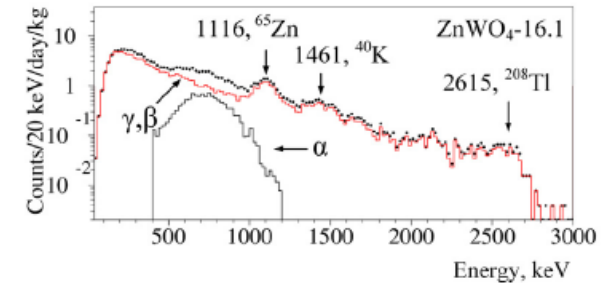


Once provided a suitable separation also at very low energy, PSD could – in principle – gives a 2nd independent but not mandatory way to exploit the directionality approach

# Radiopurity of realized ZnWO<sub>4</sub>

Measured radioactive contamination approaches that of specially developed low background DAMA-NaI(Tl):

- <1.3 μBq/kg for <sup>228</sup>Th
- ~ 0.5 ppt for <sup>232</sup>Th;
- ~ 0.2 ppt for <sup>238</sup>U;
- < 0.02 mBq/kg for <sup>40</sup>K;
- total α activity of 0.18 mBq/kg



**NIMA 935(2019)89**

**Table 2**

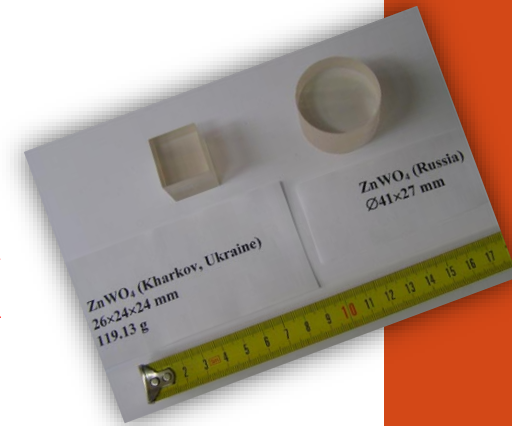
Radioactive contamination of the ZnWO<sub>4</sub> crystal scintillators. The data for one of the ZnWO<sub>4</sub> crystals studied in Ref. [25] is given for comparison. The limits are at 90% C.L.

Crystal sample	Time of measurements (h)	Activity (μBq/kg)		Counting rate [counts/(day keV kg)] in γ, β spectrum in the energy interval (MeV)	
		Total α	<sup>228</sup> Th	0.8–1.0	2.0–2.9
16.1	8097	158(3)	<0.17	0.044(1)	0.0029(1)
16.2		331(5)	<0.17	0.057(2)	0.0031(2)
16.3		577(6)	<0.17	0.082(2)	0.0030(1)
16.4		1418(9)	0.34 <sup>+0.19</sup> <sub>-0.16</sub>	0.110(2)	0.0067(2)
18.1	4299	159(8)	<1.3	0.062(4)	0.0028(3)
18.2		218(9)	<1.3	0.070(4)	0.0015(3)
ZWO-2 [25]	2130	180(30)	2(1)	0.149(3)	0.0072(4)

**Further developments are ongoing, in particular by multiple recrystallization from radio-pure part of the previous boules:  
⇒ future ZnWO<sub>4</sub> crystals with higher radio-purity expected**

# Improving radiopurity of ZnWO<sub>4</sub> crystal

- screening of zinc oxide to avoid cosmogenic <sup>65</sup>Zn
- protocol for the purification of the initial **zinc** (**vacuum distillation and filtering**) and **tungsten** (**electron beam and zone melting**)
- low-thermal gradient Czochralski technique in a platinum crucible (with very good results in producing large size crystals with high radiopurity levels)
- Segregation of radioactive elements (U, Th, Ra, K) expected (very similar compound to CdWO<sub>4</sub>) and under investigation; recrystallization could further improve radio-purity level of ZnWO<sub>4</sub>
- Detectors cut and assembled just after the growth of the crystalline bulk in a glove-box in controlled atmosphere.
- Selection of tools and abrasives for cutting and polishing the crystals
- Etc.



# DAMA-INR Kiyv measurements to study the anisotropic features of $\text{ZnWO}_4$ with alpha and neutrons

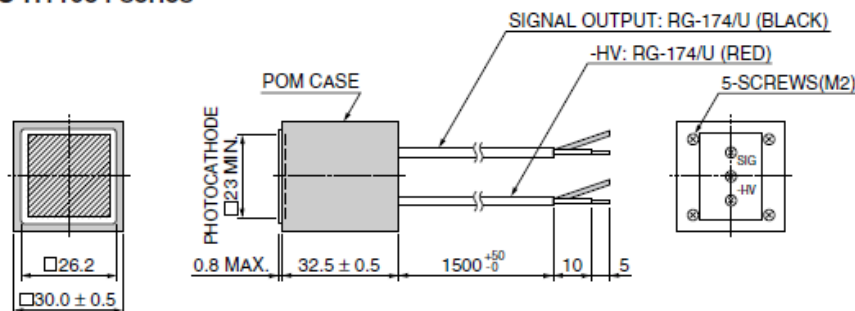
In summer 2018 new measurements using a dedicated  $\text{ZnWO}_4$  crystal started

- $\text{ZnWO}_4$  crystal =  $10.20 \times 10.20 \times 10.20 \text{ mm}^3$  (detector of reduced dimensions to investigate neutron single-scattering)
- Preliminary characterization of the crystal by using alpha particles and identification of the crystal axis

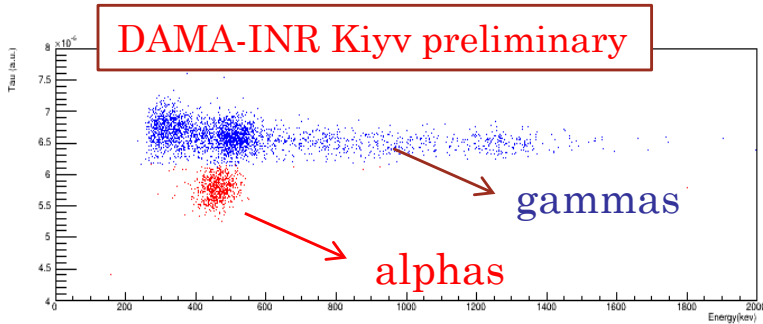
Calibration set-up:

- PMT Hamamatsu H11934-200 +  $\text{ZnWO}_4$
- LeCroy Oscilloscope 24Xs-A, 2.5 Gs/s, 200MHz band width
- Metallic Box
- Pulse profiles acquired in a time window of  $100 \mu\text{s}$

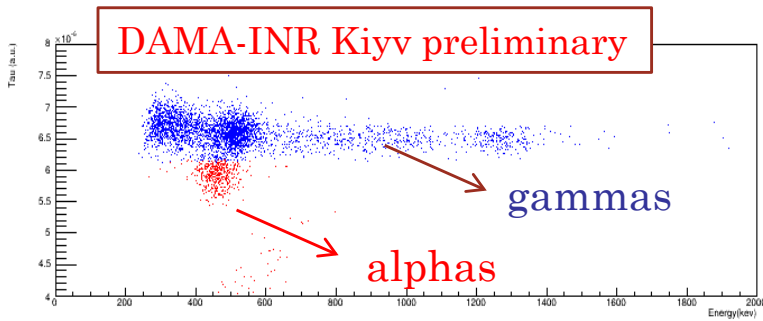
## ● H11934 series



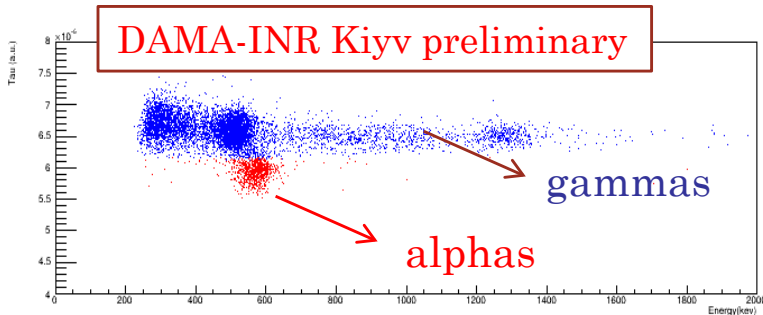
We performed measurements irradiating contemporaneously the crystal with a  $\gamma$  source ( $^{22}\text{Na}$ ) and a collimated alpha source ( $^{241}\text{Am}$ ) along the three crystal axes (P1, P2, P3)



P2



P1

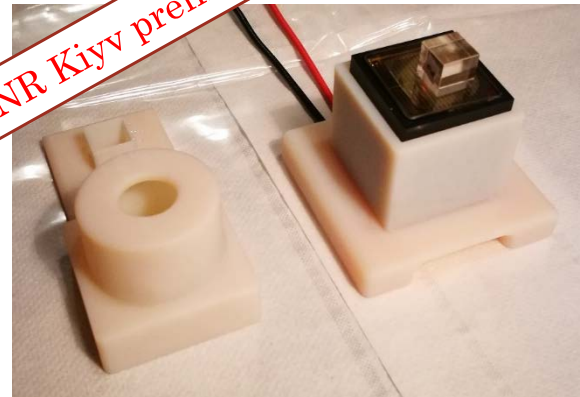


P3

About  $\alpha$  source:

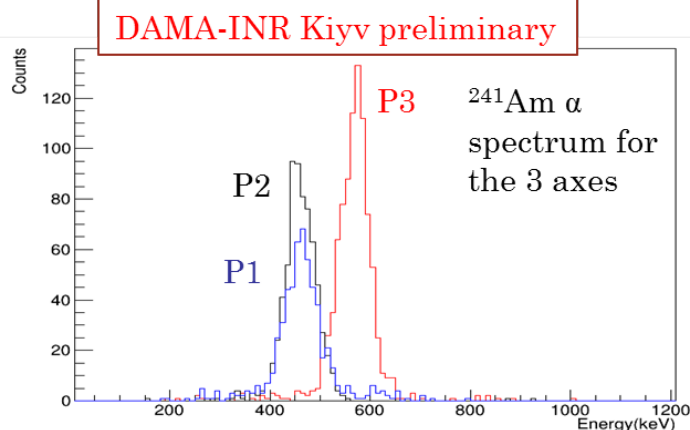
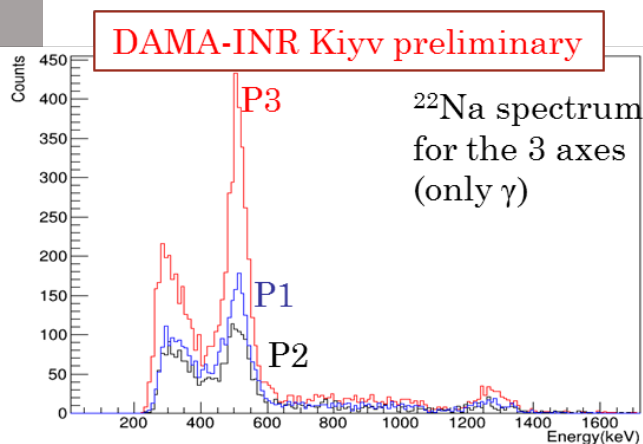
- calibration with Si detector to measure the energy of the  $\alpha$  emitted by the used  $^{241}\text{Am}$  source
- comparison of the obtained energy distribution with the one obtained by considering a reference electrodeposited  $^{241}\text{Am}$  source (measurements show no energy degradation for the alpha emitted by the used source)

DAMA-INR Kiyv preliminary





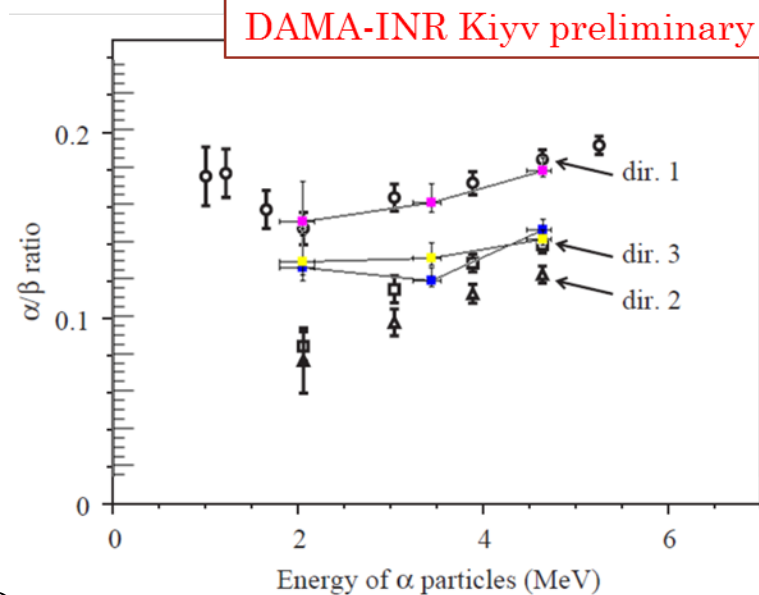
# DAMA-INR Kiyv measurements to identify the $ZnWO_4$ crystal axis by using $\alpha$ particles



Identified the axis with highest light response (**P3**)

$E_\alpha$ (MeV)	Axes	$\alpha/\beta$	$P_x/P_3$	$P_3/P_x$
2,04	P1	$0,126^{+0,18}_{-0,07}$	0,83	1,20
	P2	$0,129^{+0,18}_{-0,07}$	0,84	1,17
	P3	$0,151^{+0,21}_{-0,08}$	1,00	1,00
3,43	P1	$0,119^{+0,07}_{-0,03}$	0,74	1,35
	P2	$0,131^{+0,08}_{-0,04}$	0,81	1,23
	P3	$0,161^{+0,10}_{-0,05}$	1,00	1,00
4,63	P1	$0,146^{+0,06}_{-0,03}$	0,82	1,22
	P2	$0,141^{+0,05}_{-0,03}$	0,80	1,25
	P3	$0,177^{+0,07}_{-0,03}$	1,00	1,00

DAMA-INR Kiyv preliminary



❑  $\alpha/\beta$  discrimination capability of  $ZnWO_4$

❑  $\alpha/\beta$  ratio at  $\approx 2$  MeV:  $(\alpha/\beta)_{P3} \approx 0.15$ ,  $(\alpha/\beta)_{P1} \approx (\alpha/\beta)_{P2} \approx 0.13$

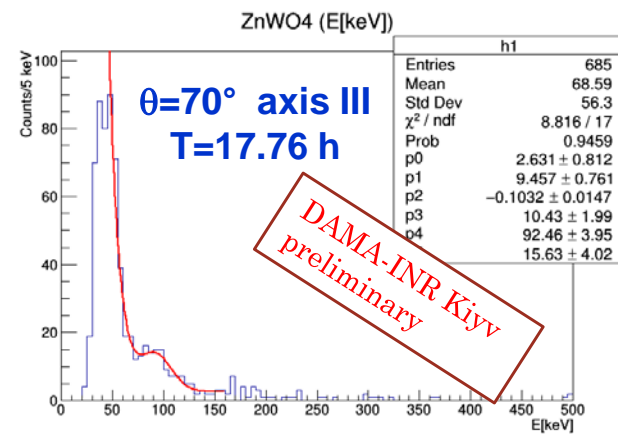
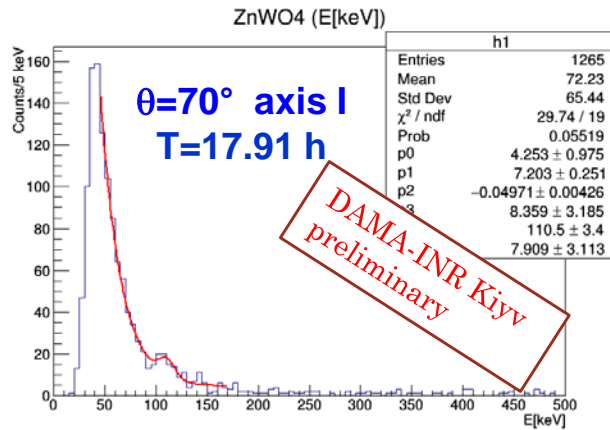
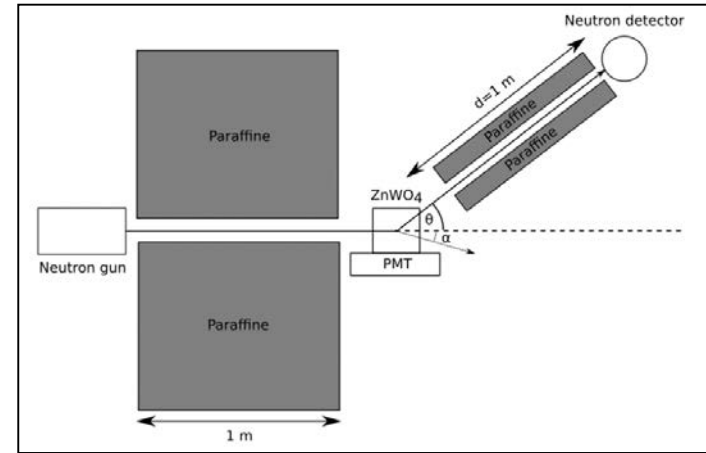
# First measurement of anisotropy for low energy nuclear recoils in $\text{ZnWO}_4$

DAMA-INR Kiyv preliminary – work in progress

MP320 Neutron Generator ( $E_n = 14 \text{ MeV}$ )  
 @ ENEA-Casaccia lab ( $D + T \rightarrow n + {}^4\text{He}$ )

Scattering angle of neutron [deg]: 70.0000

Nucl.	Target mass [GeV]	$E_{\text{recoil}}$ [keV]	$\alpha$ lab. target
Zn	60.9	281.4	54.6
W	171.2	100.7	54.9
O	14.9	1115.7	53.3

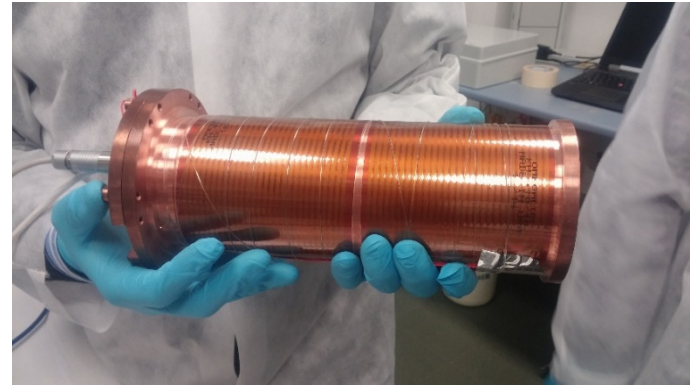


DAMA-INR Kiyv preliminary

Angle	Axis	$E_{\text{peak}}$ (keVee)	$E_{\text{recoil,O}}$ (keV)	Q	$Q_I / Q_{III}$
70	I	110.5±3.4	1116	0.099±0.003	1.20±0.06
70	III	92.5±3.9	1116	0.083±0.003	

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

- ❑ Cryostat for low temperature measurement with scintillation detectors realized
- ❑ Test of the Cryostat in progress
- ❑ Lowering the energy threshold (new PMT with higher QE, SiPM, APD, SDD, ...)
- ❑ Measurements of anisotropy at low energy with MP320 Neutron Generator ( $E_n = 14$  MeV) at Casaccia lab
- ❑ New data taking with new detector with improved features and identified crystal axis
- ❑ Improved setup and experimental size

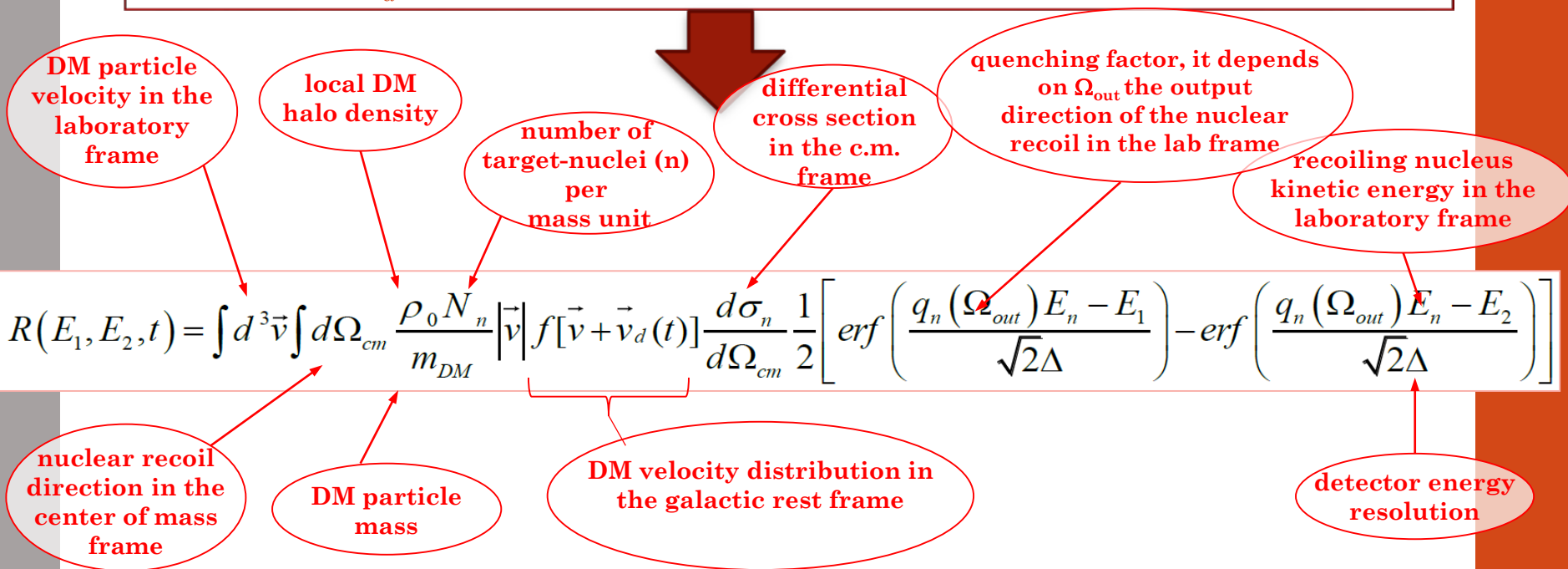


# Signal rate in a given scenario

Eur. Phys. J. C 73 (2013) 2276

Because of the *light response anisotropy for heavy particles* recoil nuclei induced by the considered DM candidates could be discriminated from the background thanks to the *expected variation of their low energy distribution along the day*

The expected signal counting rate in the energy window  $(E_1, E_2)$  is a function of the time  $t$  ( $v_d(t)$  the detector velocity in the galactic rest frame)



NB: Many quantities are model dependent and a model framework has to be fixed: in this example, for simplicity, a set of assumptions and of values have been fixed, without considering the effect of the existing uncertainties on each one of them and without considering other possible alternatives

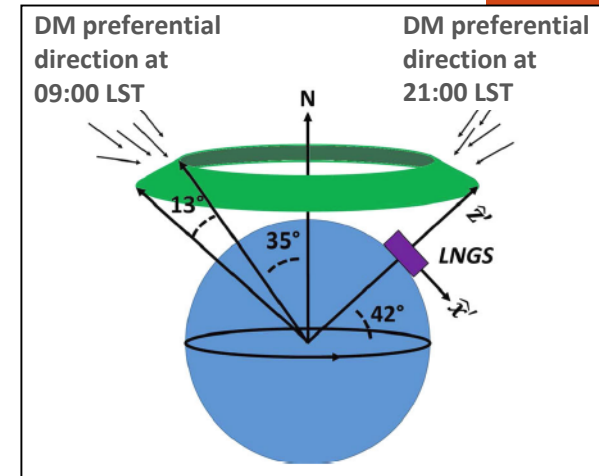
# Example of expected signal

It is very convenient to consider an experiment performed at the LNGS latitude ( $42^{\circ}27'N$ )

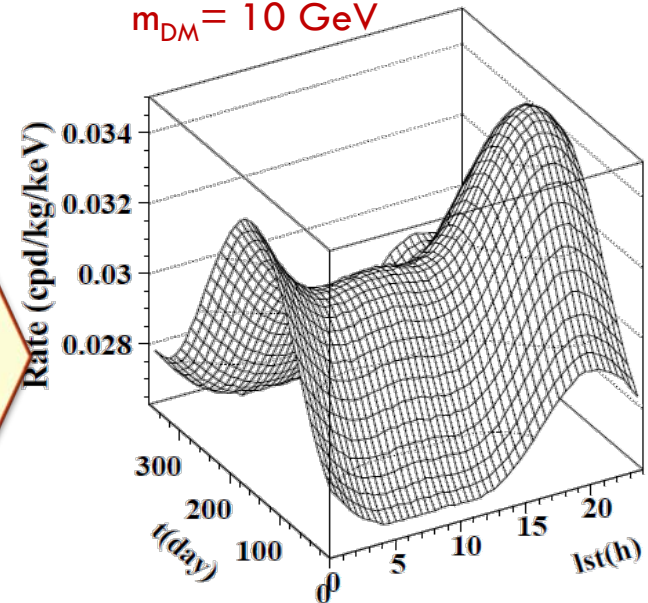
⇒ at 21:00 h LST the DM particles come mainly from the top, and 12 h later from the North and parallel to the horizon line

If we arrange the  $ZnWO_4$  crystal axis so that:

- The one with the largest light output is vertical and
  - the one with the smallest light output points north
- ⇒ range of variability of the anisotropic detector response during a sidereal day is at maximum



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



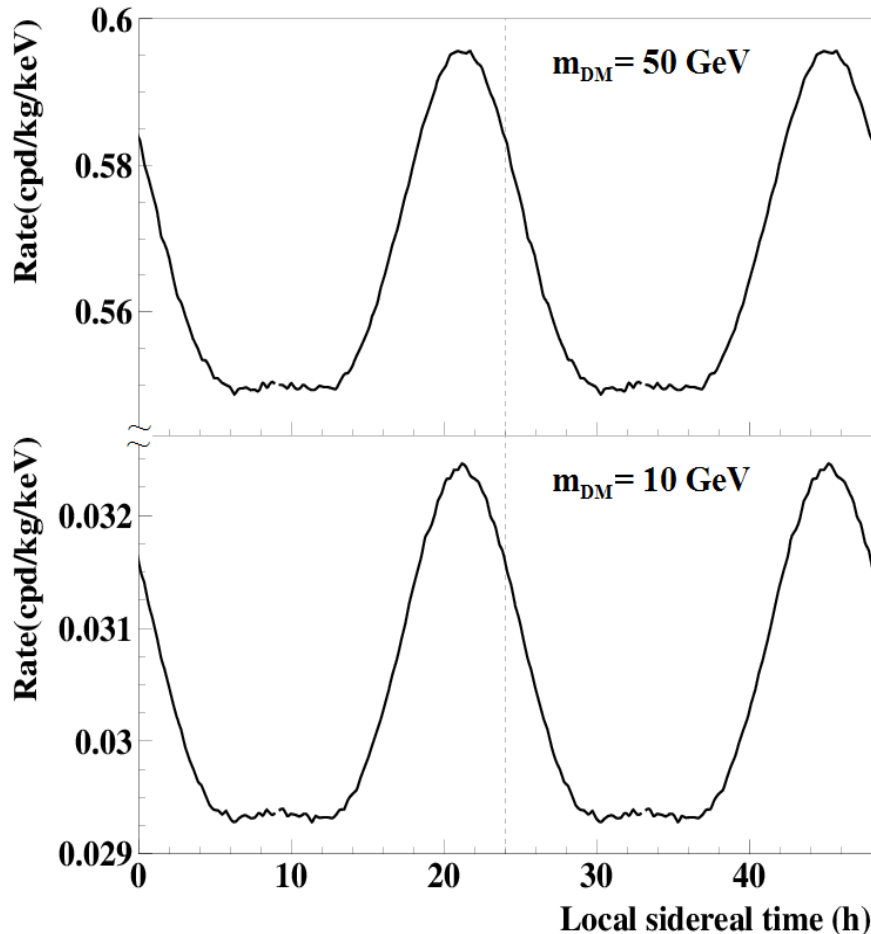
The diurnal effect refer to the sidereal day and not to the solar day

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)

TEST: Identical sets of crystals placed in the same set-up with different axis orientation will observe consistently different time evolution of the rate

# Example of expected signal rate in the given scenario

Signal rate in [2-3] keV energy range with  $\xi\sigma_p = 5 \times 10^{-5}$  pb



- Maximum rate at 21 h sidereal time of LNGS, when the DM preferential arrival direction is near the zenith, that is near the crystal axis with the largest light output.
- Analogous results can be obtained also analysing the anisotropic behaviour of the pulse shape of scintillation events.

**The signature is very distinctive**

# Conclusions

- Anisotropic  $\text{ZnWO}_4$  detectors are promising detectors to investigate the directionality for those DM candidates inducing just nuclear recoils
- The use of anisotropic scintillators allows us to overcome limitations due to track measurement
- With respect to the positive DM annual modulation results obtained by DAMA, such an experiment (ADAMO - about 200 kg of  $\text{ZnWO}_4$ , 5 years of data taking with energy threshold at 2 keV and a background counting rate  $<10^{-2}$  cpd/kg/keV) can obtain, with a completely different approach, complementary information on the nature and interaction type of the DM candidate

**THANKS FOR YOUR  
ATTENTION**