DAMA, INR-Kyiv,
ENEA-Casaccia

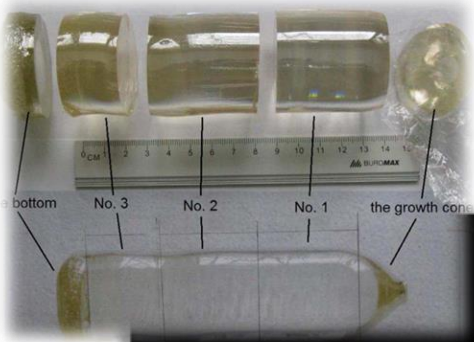
Development of $ZnWO_4$ crystal scintillators for rare events search

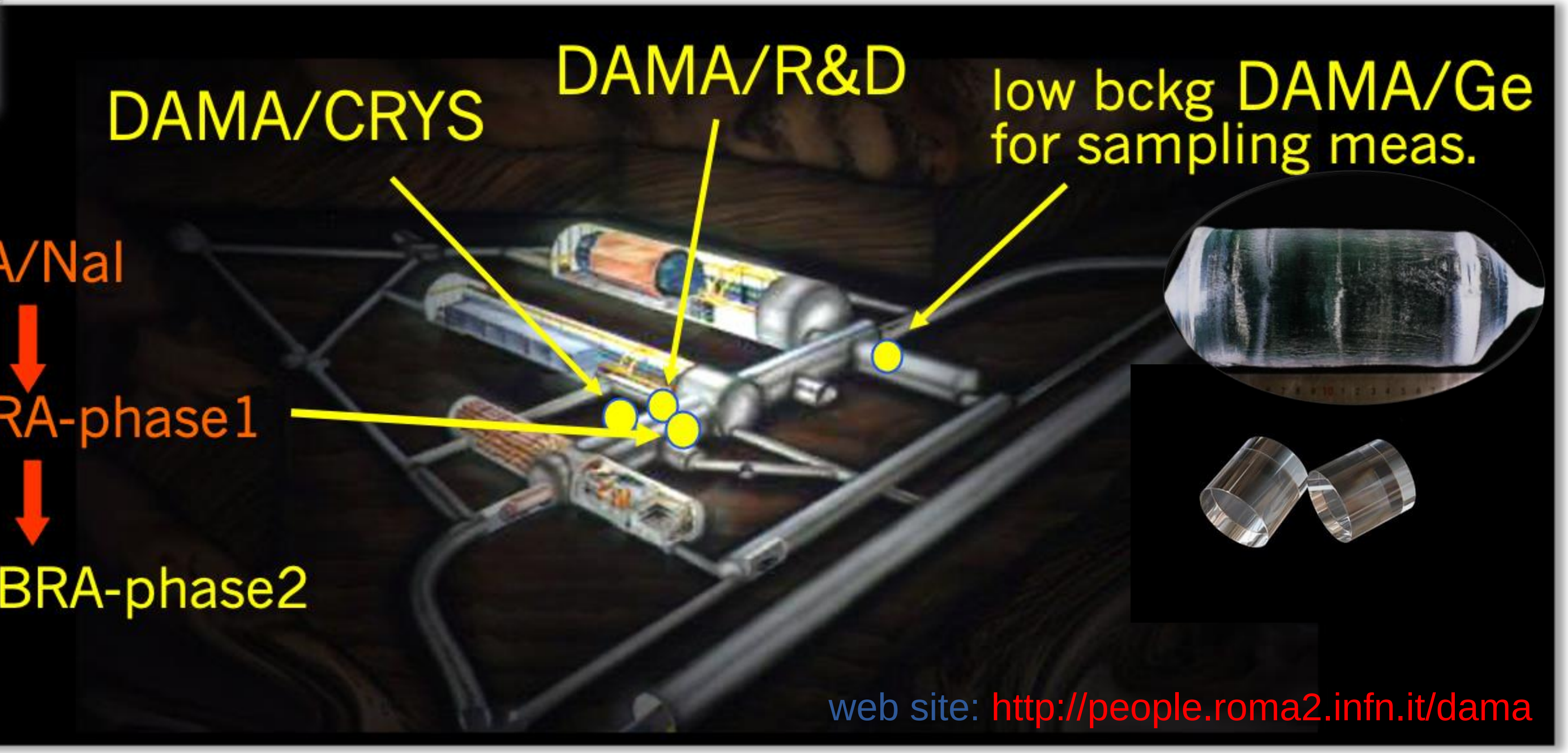
ICNFP 2021
Kolymbari, Crete, Greece
23 August to 2 September 2021

INFN
Fabio Cappella
INFN Roma

DAMA

an observatory for rare processes @ LNGS





DAMA/CRYS

DAMA/R&D

low bckg DAMA/Ge for sampling meas.

DAMA/NaI
↓
DAMA/LIBRA-phase1
↓
DAMA/LIBRA-phase2

web site: http://people.roma2.infn.it/dama

Roma Tor Vergata, Roma La Sapienza, LNGS, IHEP/Beijing

+ by-products and small scale expts.: INR-Kiev + other institutions

+ neutron meas.: ENEA-Frascati, ENEA-Casaccia

+ in some studies on ββ decays (DST-MAE and Inter-Universities project): IIT Kharagpur and Ropar, India

Rare events search with zinc tungstate ($ZnWO_4$) detectors

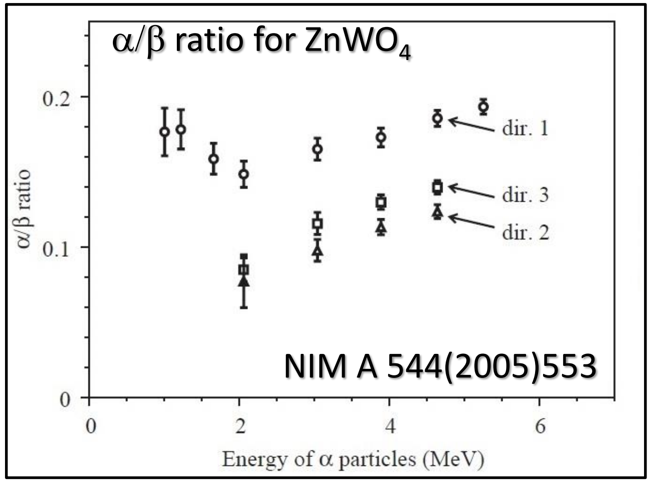
1. Double Beta decay in Zn and W nuclides and α decay of ^{180}W

   $ZnWO_4$ scintillators contain four potentially $\beta\beta$ active isotopes: ^{64}Zn, ^{70}Zn, ^{180}W and ^{186}W

Transition	Energy release ($Q_{\beta\beta}$) (keV)	Isotopic abundance (%)	Decay channels	Number of mother nuclei in 100 g of $ZnWO_4$ crystal
$^{64}Zn \rightarrow {}^{64}Ni$	1095.7(0.7)	49.17(75)	2ε, $\varepsilon\beta^+$	9.45×10^{22}
$^{70}Zn \rightarrow {}^{70}Ge$	998.5(2.2)	0.61(10)	$2\beta^-$	1.17×10^{21}
$^{180}W \rightarrow {}^{180}Hf$	144(4)	0.12(1)	2ε	2.31×10^{20}
$^{186}W \rightarrow {}^{186}Os$	489.9(1.4)	28.43(19)	$2\beta^-$	5.47×10^{22}

2. Study of the Dark Matter (DM) particle component in the galactic halo with the directionality approach

   Thanks to the anisotropic features of the $ZnWO_4$ crystal scintillators, the light response and the pulse shape induced by heavy ionizing particles (p, α, nuclear recoils) depend on their direction wrt the crystal axes



α/β ratio for $ZnWO_4$

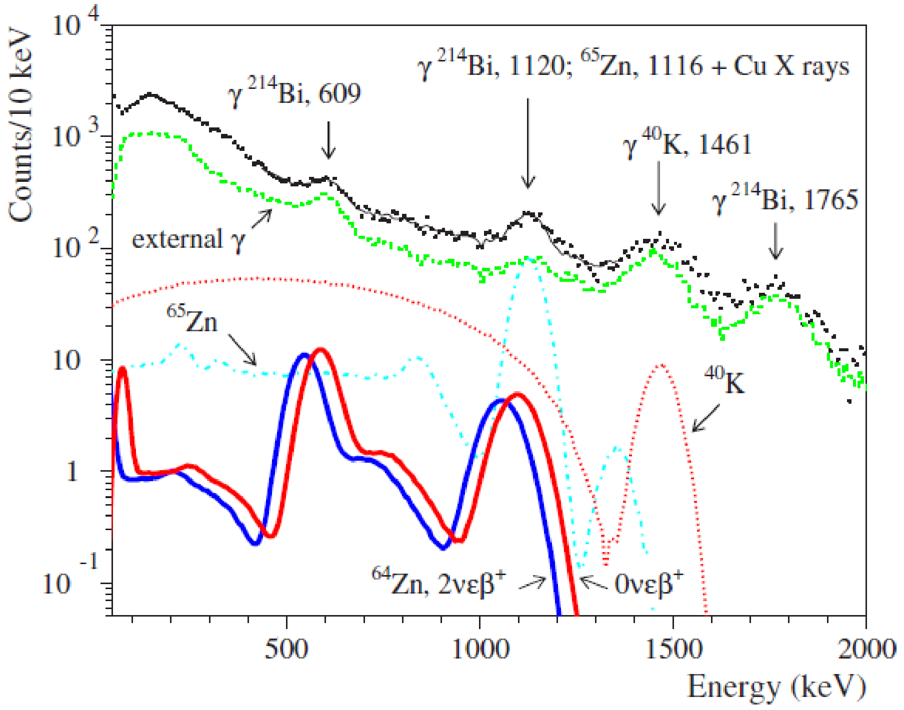
NIM A 544(2005)553

Search for ββ decay with $ZnWO_4$ crystal scintillators

J.Phys.G 38 (2011) 115107

A search for the ββ decay of zinc and tungsten isotopes was performed by DAMA + INR-Kyiv in 2011 with the help of radiopure $ZnWO_4$ crystal scintillators (0.1–0.7 kg) at LNGS (exposure: 0.529 kg×yr)

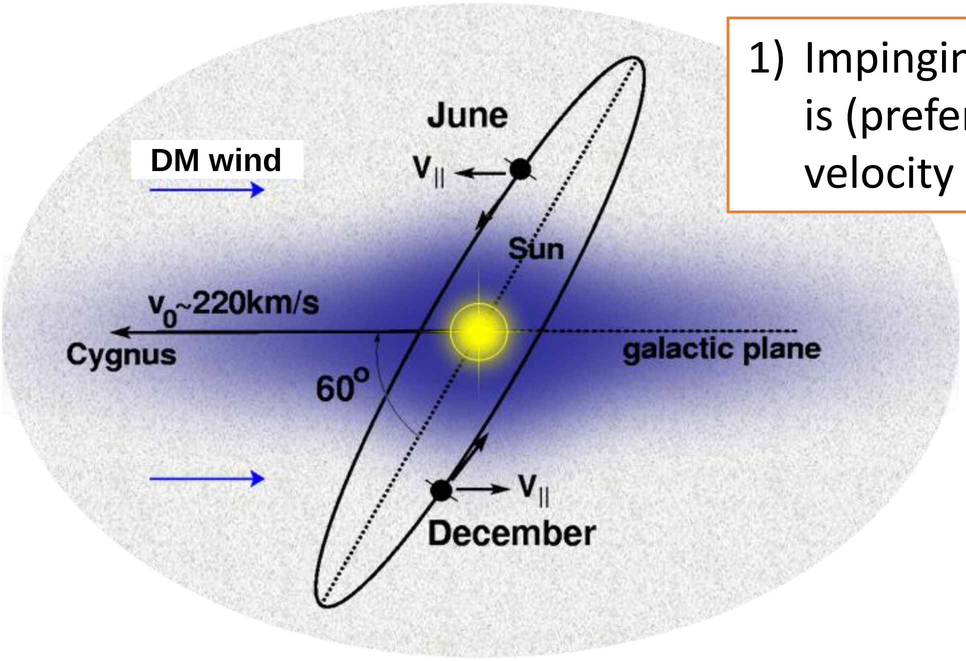
Transition	Decay channel	Level of the daughter nucleus	Experimental limits on $T_{1/2}$, yr at 90% CL		Theoretical estimations of the half-lives $T_{1/2}$, yr
			Present work	The best previous results	($\langle m_\nu \rangle = 1$ eV for $0\nu 2\beta$ decay)
$^{64}Zn \to {}^{64}Ni$	$2\nu 2K$	g.s.	$\geqslant 1.1 \times 10^{19}$	$\geqslant 6.0 \times 10^{16}$ [54]	$(1.9\text{–}7.1) \times 10^{26}$ [56] $(1.2 \pm 0.2) \times 10^{25}$ [57]
	$0\nu 2\varepsilon$	g.s.	$\geqslant 3.2 \times 10^{20}$	$\geqslant 7.4 \times 10^{18}$ [55]	–
	$2\nu\varepsilon\beta^+$	g.s.	$\geqslant 9.4 \times 10^{20}$	$= (1.1 \pm 0.9) \times 10^{19}$ [58] $\geqslant 1.3 \times 10^{20}$ [59]	$(0.9\text{–}2.2) \times 10^{35}$ [56] $(4.7 \pm 0.9) \times 10^{31}$ [57]
	$0\nu\varepsilon\beta^+$	g.s.	$\geqslant 8.5 \times 10^{20}$	$\geqslant 1.3 \times 10^{20}$ [59]	–
$^{70}Zn \to {}^{70}Ge$	$2\nu 2\beta^-$	g.s.	$\geqslant 3.8 \times 10^{18}$	$\geqslant 1.3 \times 10^{16}$ [46]	$4.5 \times 10^{21} - 3.6 \times 10^{24}$ [60] $2.5 \times 10^{21} - 6.4 \times 10^{23}$ [61] 7.0×10^{23} [56] $\geqslant 3.1 \times 10^{22}$ [62]
	$0\nu 2\beta^-$	g.s.	$\geqslant 3.2 \times 10^{19}$	$\geqslant 7.0 \times 10^{17}$ [46]	9.8×10^{25} [60]
	$0\nu 2\beta^- M1$	g.s.	$\geqslant 6.0 \times 10^{18}$	–	–
	$0\nu 2\beta^- M2$	g.s.	$\geqslant 4.7 \times 10^{18}$	–	–
	$0\nu 2\beta^- bM$	g.s.	$\geqslant 5.4 \times 10^{18}$	–	–
$^{180}W \to {}^{180}Hf$	$2\nu 2K$	g.s.	$\geqslant 1.0 \times 10^{18}$	$\geqslant 7.0 \times 10^{16}$ [30]	–
	$0\nu 2\varepsilon$	g.s.	$\geqslant 1.3 \times 10^{18}$	$\geqslant 9.0 \times 10^{16}$ [30]	$2.5 \times 10^{24} - 2.5 \times 10^{26}$ [23] $3.3 \times 10^{27} - 5.0 \times 10^{30}$ [24] $3.0 \times 10^{22} - 4.0 \times 10^{27}$ [25]
$^{186}W \to {}^{186}Os$	$2\nu 2\beta^-$	g.s.	$\geqslant 2.3 \times 10^{19}$	$\geqslant 3.7 \times 10^{18}$ [30]	$7.1 \times 10^{23} - 1.2 \times 10^{25}$ [60] $\geqslant 6.1 \times 10^{24}$ [19]
	$2\nu 2\beta^-$	2^+_1(137 keV)	$\geqslant 1.8 \times 10^{20}$	$\geqslant 1.0 \times 10^{19}$ [30]	–
	$0\nu 2\beta^-$	g.s.	$\geqslant 1.0 \times 10^{21}$	$\geqslant 1.1 \times 10^{21}$ [30]	6.4×10^{24} [60]
	$0\nu 2\beta^-$	2^+_1(137 keV)	$\geqslant 9.0 \times 10^{20}$	$\geqslant 1.1 \times 10^{21}$ [30]	–
	$0\nu 2\beta^- M1$	g.s.	$\geqslant 5.8 \times 10^{19}$	$\geqslant 1.2 \times 10^{20}$ [30]	–
	$0\nu 2\beta^- M2$	g.s.	$\geqslant 1.1 \times 10^{19}$	–	–
	$0\nu 2\beta^- bM$	g.s.	$\geqslant 1.1 \times 10^{19}$	–	–



Few other nuclides among 34 candidates to 2ε, εβ⁺ and 2β⁺ processes were studied at a similar level of sensitivity in direct experiments

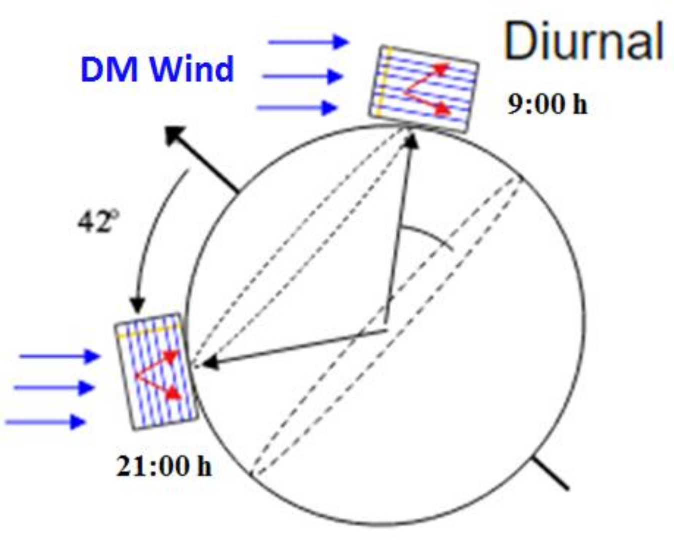
The directionality approach

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



1) Impinging direction of DM particle is (preferentially) opposite to the velocity of the Sun in the Galaxy

2) 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



3) The direction of the induced nuclear recoil is strongly correlated with that of the impinging DM particle
4) 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

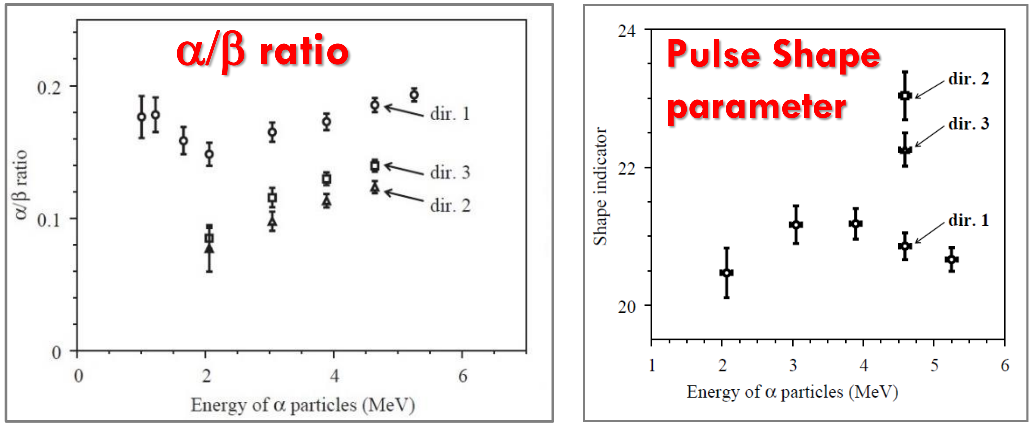
Directionality with anisotropic scintillators

Firstly proposed in [P. Belli et al., Il Nuovo Cim. C 15 (1992) 475; R. Bernabei et al., EPJC28(2003)203]

Anisotropic Scintillators:

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

An example: α Particles in $ZnWO_4$



NIM A 544(2005)553

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 nuclear recoils

Comparison with directionality experiments using Low Pressure Time Projection Chambers (TPC):

- **TPC advantages:** the range of recoiling nuclei is of the order of mm (while it is ~μm in solid detectors)
- **TPC drawbacks:** γ rejection and high angular and spatial resolution is required at very low energy; limitations on mass and stability

Other techniques under development:

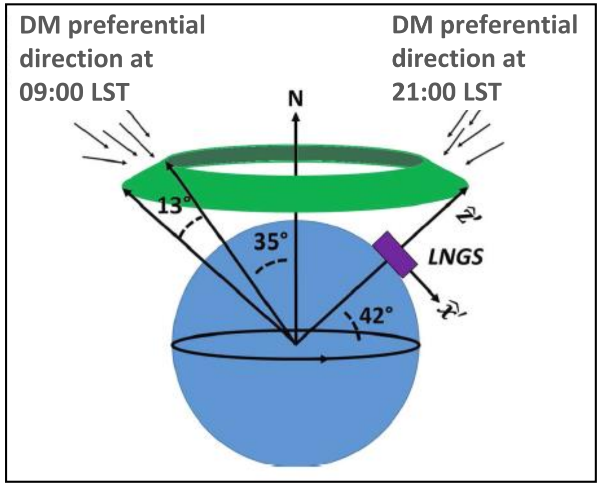
Nuclear Emulsions, DNA, Diamonds, Carbon nanotubes based detector, Columnar Recombination in LAr/LXe-TPC

Example of expected signal

It is very convenient to consider an experiment performed at the LNGS latitude (**42°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:

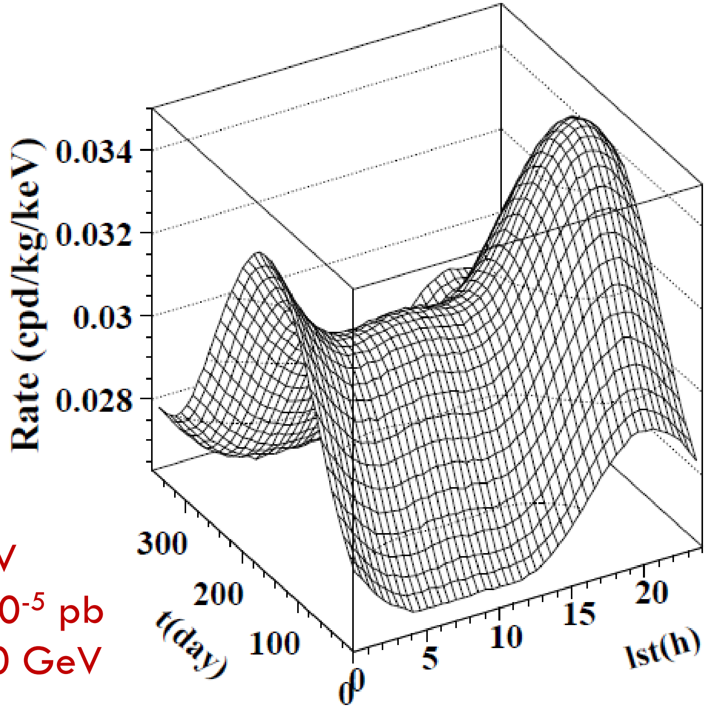
- The one with the largest light output is vertical and
- the one with the smallest light output points north

⇒ range of variability of the anisotropic detector response during a sidereal day is at maximum

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

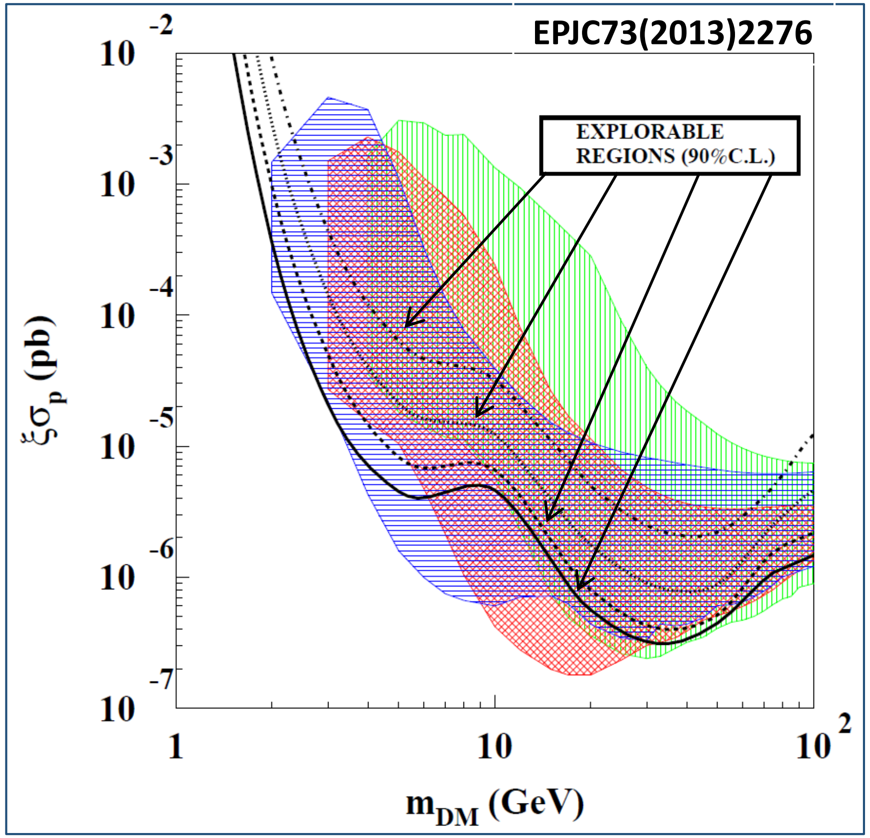


[2-3] keV
$\sigma_p = 5\times10^{-5}$ pb
$m_{DM} = 10$ GeV

ADAMO project: example of reachable sensitivity in a given scenario

Assumptions:

- simplified model framework
- 200 kg of $ZnWO_4$
- 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 —·—·—·—



EPJC73(2013)2276

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 and blue) obtained with a corollary analysis of the 9.3σ C.L. model independent result of DAMA/NaI + DAMA/LIBRA-phase1 in terms of scenarios for the DM candidates considered here
(updated DAMA allowed regions are reported in NPAE 20 (2019) 317)

Development of $ZnWO_4$ crystal scintillators

$ZnWO_4$ anisotropic scintillator: a very promising detector

[NIMA 544 (2005) 553, Eur. Phys. J. C 73 (2013) 2276]

- ✓ Very good anisotropic features
- ✓ High level of radiopurity
- ✓ 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 ~ kg masses feasible

Density (g/cm^3)	*7.87*
Melting point (ºC)	*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 (µs)	*24*

The main ongoing R&Ds and studies (ADAMO project):

- ➢ Further increase the radio-purity level
- ➢ Improve the optical properties
- ➢ Increase the light yield to further decrease the energy threshold (Study the light yield response vs the operation temperature)
- ➢ Study the anisotropies property at energy of interest for DM particle nuclear recoils

JINST15(2020)07,C07037; JINST15(2020)05,C05055; NIMA935(2019)89; NIMA833(2016)77; JPCS718(2016)4,042011; EPJC73(2013)2276; NIMA626-627(2011)3; JP38(2011)115107 NPA826(2009)256; PLB658(2008)193

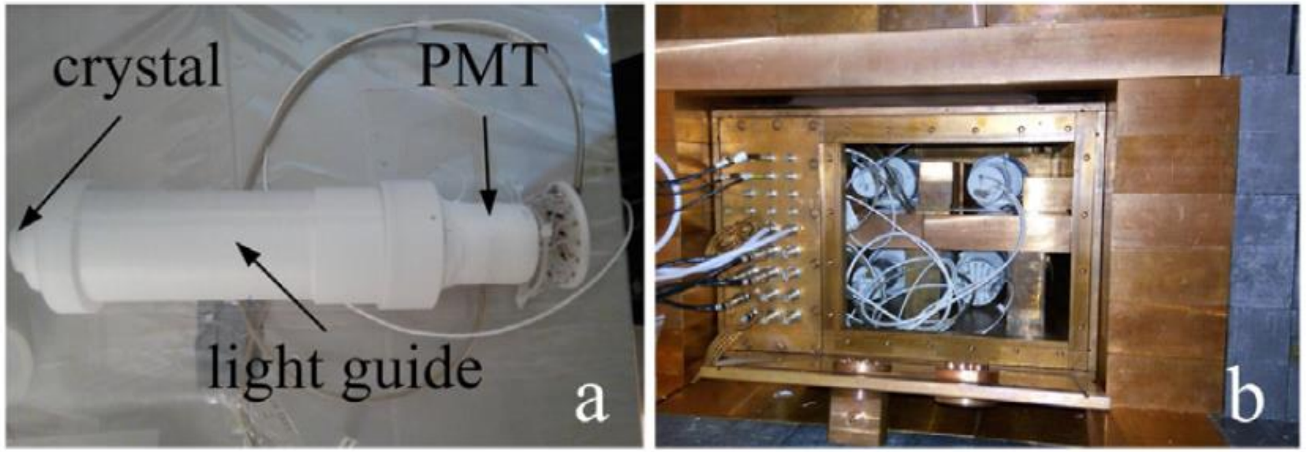
R&D of radiopure high quality $ZnWO_4$ crystal scintillators

Radiopurity of $ZnWO_4$ studied with several crystal samples, and strategies in low-background set-ups at LNGS:

- screening of zinc oxide to avoid cosmogenic ^{65}Zn
- protocol for the purification of the initial **zinc** (vacuum distillation and filtering) and **tungsten** (electron beam zone melting)
- low-thermal gradient Czochralski technique in a platinum crucible (very good results for large size crystals with high radiopurity levels)
- Recrystallization: segregation of radioactive elements is expected as in $CdWO_4$ (very similar compound)
- 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

⇒ $ZnWO_4$ is one of the most radio-pure crystal scintillator, the best results obtained for the main contaminants are:

- < 0.17 μBq/kg for ^{228}Th (~0.04 ppt of ^{232}Th)
- < 2 μBq/kg for ^{226}Ra (~ 0.2 ppt for ^{238}U)
- < 20 μBq/kg for ^{40}K (0.6 ppb ^{nat}K)
- Total α activity: 0.16 mBq/kg



The recent R&D substantially improved the optical properties of the material

Next step: development and the test of large-volume $ZnWO_4$ crystal scintillators

[NIM A 935 (2019) 89]
[NIM A 626-627 (2011) 31]

Measurements of $ZnWO_4$ anisotropic response to nuclear recoils for the ADAMO project

Eur.Phys.J.A 56 (2020) 83

- In summer 2018 a campaign of measurements using a dedicated $ZnWO_4$ crystal to study the anisotropic features of the detector for low energy nuclear recoils started
- Preliminary measurements with a collimated α source have been performed
- After α calibrations a campaign of measurements at ENEA-Casaccia with a 14 MeV neutron beam has been carried out


Very transparent and radio-pure

$ZnWO_4$ crystal = 10 x 10 x 10 mm^3
(detector of reduced dimensions to investigate neutron single-scattering)

Studying the response of the $ZnWO_4$ to ^{241}Am α source

Eur.Phys.J.A 56 (2020) 83

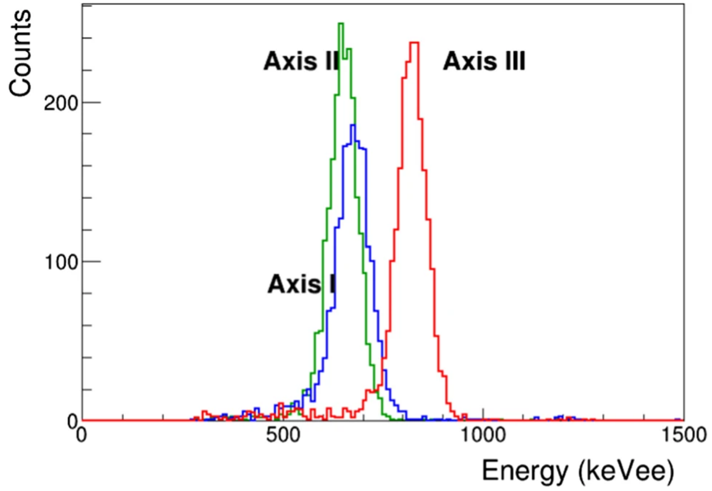
Calibration set-up:

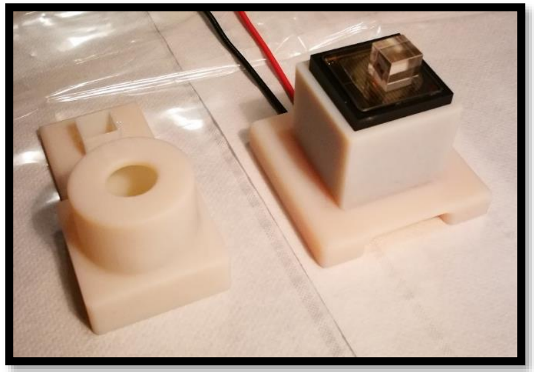
- PMT Hamamatsu H11934-200 (transit time ≈ 5 ns) + $ZnWO_4$
- LeCroy Oscilloscope 24Xs-A, 2.5 Gs/s, 200MHz bandwidth
- Pulse profiles acquired in a time window of 100 μs

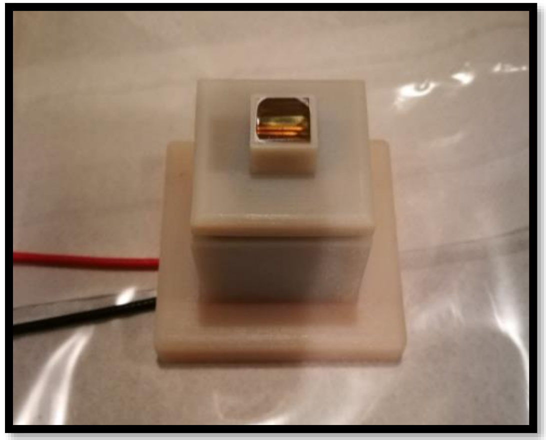
Crystal irradiated contemporaneously with γ (^{22}Na) and α (^{241}Am) sources along the three crystal axes

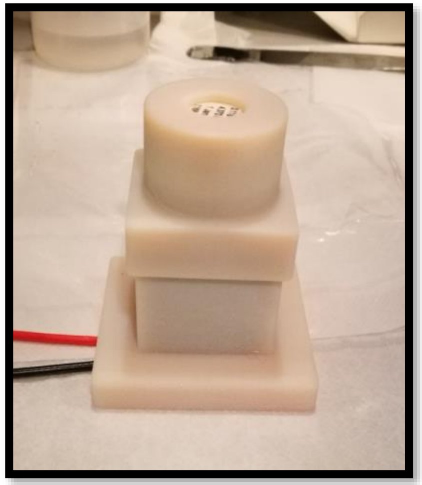
Different α energies obtained with Mylar foils and measured with Si detector

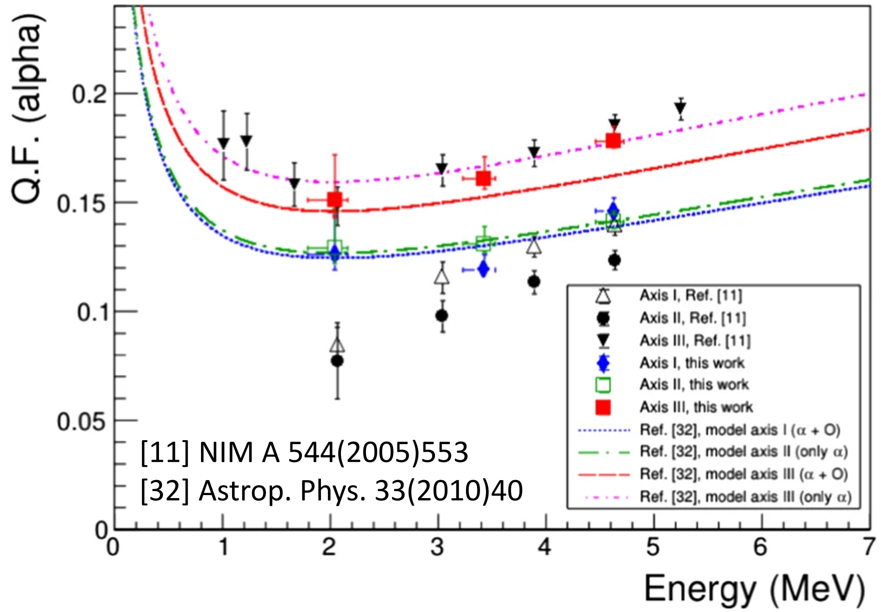
Very efficient PSD capability to discriminate α and γ











[11] NIM A 544(2005)553

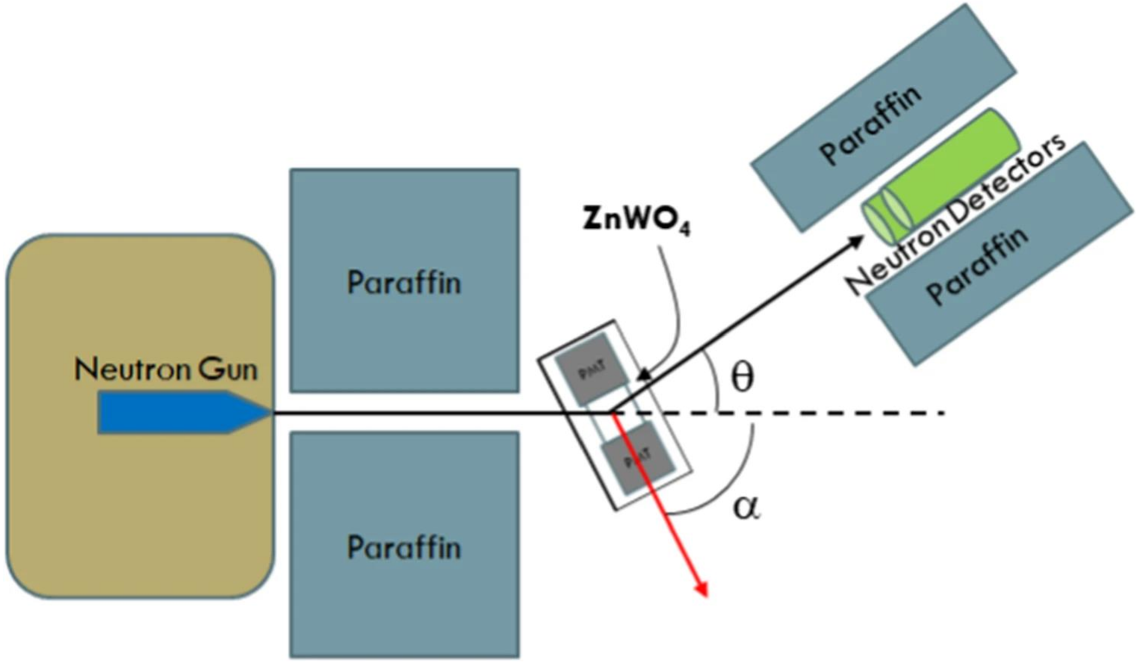
[32] Astrop. Phys. 33(2010)40

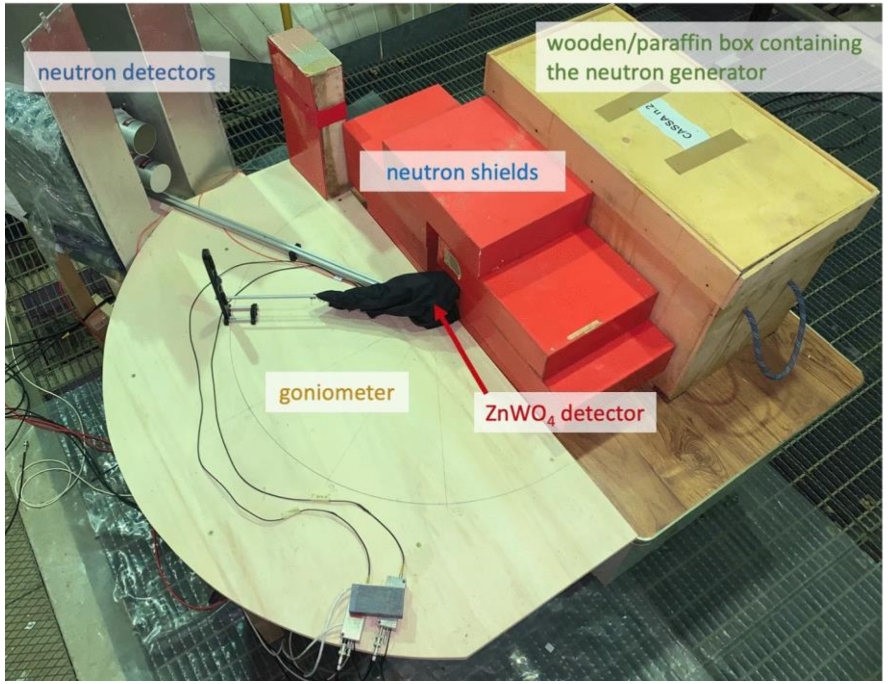
Studying the response of the $ZnWO_4$ with a neutron gun

Eur.Phys.J.A 56 (2020) 83

- Set-up:
  - ✓ $ZnWO_4$ Crystal (10 x 10 x 10 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 $ZnWO_4$ occurred in a well defined time window (TOF)
- Once fixed the θ angle, the recoil direction and energy are fixed
- Measurements performed at different θ angles and for different $ZnWO_4$ orientation (nuclear recoils along axes III and I)





Response of $ZnWO_4$ to neutrons: data analysis

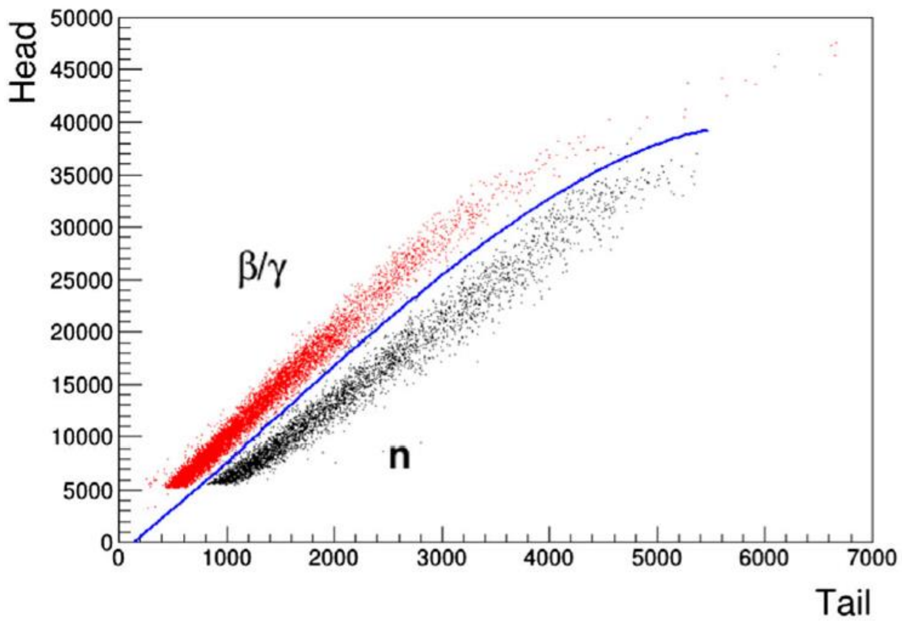
Eur.Phys.J.A 56 (2020) 83

Considered scattering angles: θ = 60°, 70°, 80°

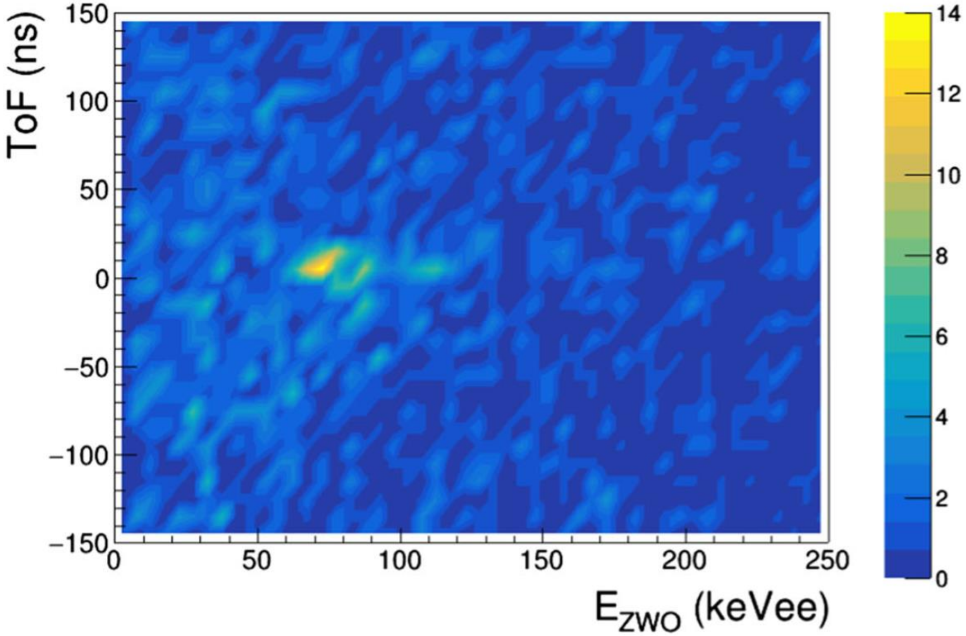
The used software energy threshold E_{th}=30 keVee allows the observation of the O recoils (Zn and W expected between 7 and 14 keVee and between 1.6 and 2.5 keVee, respectively)

Selection of the events due to elastic scattering of neutrons in the $ZnWO_4$ crystal:

- PSD analysis to select neutron induced events in the neutron detectors
- Study of the TOF between the $ZnWO_4$ detector and the neutron detectors



Example of γ/n separation by PSD in EJ-309 liquid scintillator with head/tail analysis



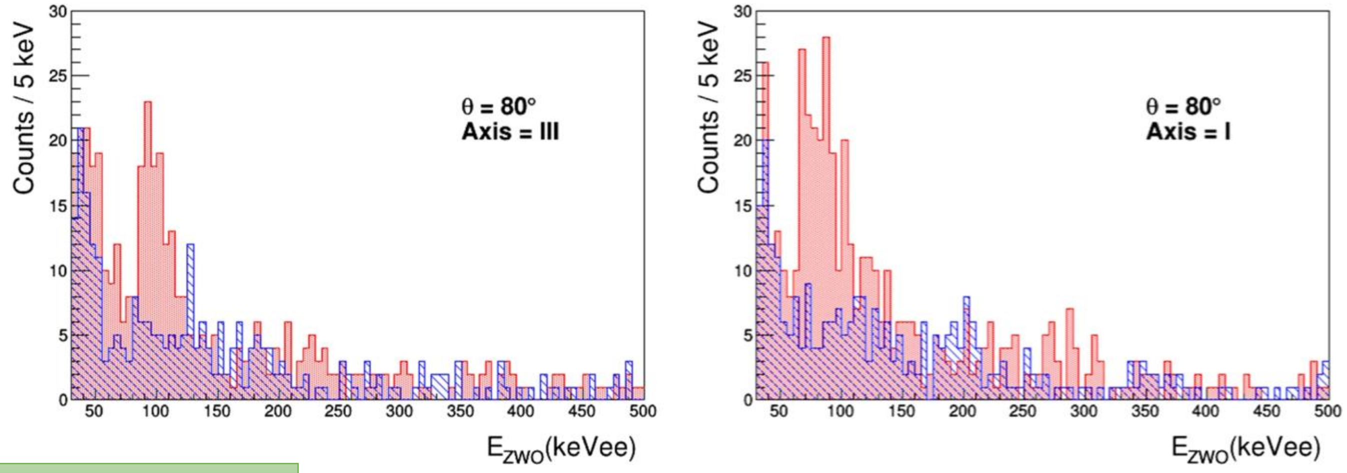
Examples of the bi-dimensional plot *TOF* vs $ZnWO_4$ energy for coincidences obtained for the case of θ = 70° and axis I

Response of $ZnWO_4$ to neutrons: results

Eur.Phys.J.A 56 (2020) 83

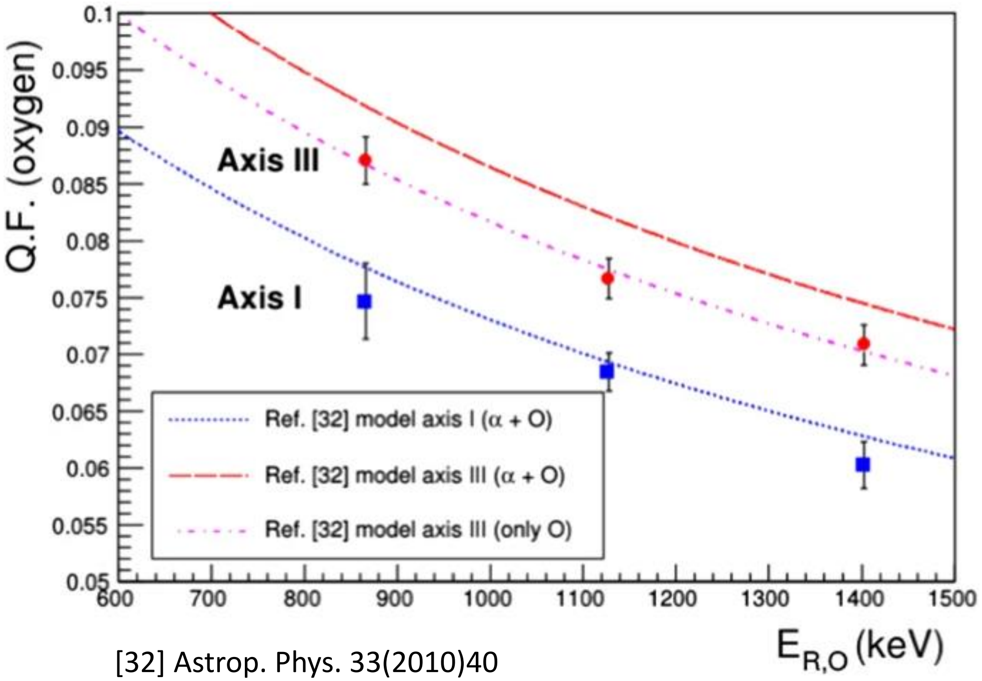
Energy distributions in $ZnWO_4$ for coincidence events when neutrons are identified in EJ-309 and two *TOF* windows are considered (case θ=80°):

- Events with proper *TOF* for **neutron induced recoils**
- Events with off-window *TOF:* **random coincidences**



First evidence for anisotropy on the response of $ZnWO_4$ for nuclear recoils

θ	Crystal axis	E_{ZWO} (keVee)	σ (keVee)	$E_{R,O}$ (keV)	Quenching factor, Q	Q_{III}/Q_I
80°	III	99.3 ± 2.5	9	1402	0.0708 ± 0.0018	1.174 ± 0.051
	I	84.5 ± 2.9	12		0.0603 ± 0.0021	
70°	III	86.5 ± 2.0	7	1128	0.0767 ± 0.0018	1.121 ± 0.038
	I	77.2 ± 1.9	10		0.0684 ± 0.0017	
60°	III	75.4 ± 1.8	9	866	0.0871 ± 0.0021	1.166 ± 0.059
	I	64.7 ± 2.9	10		0.0747 ± 0.0033	

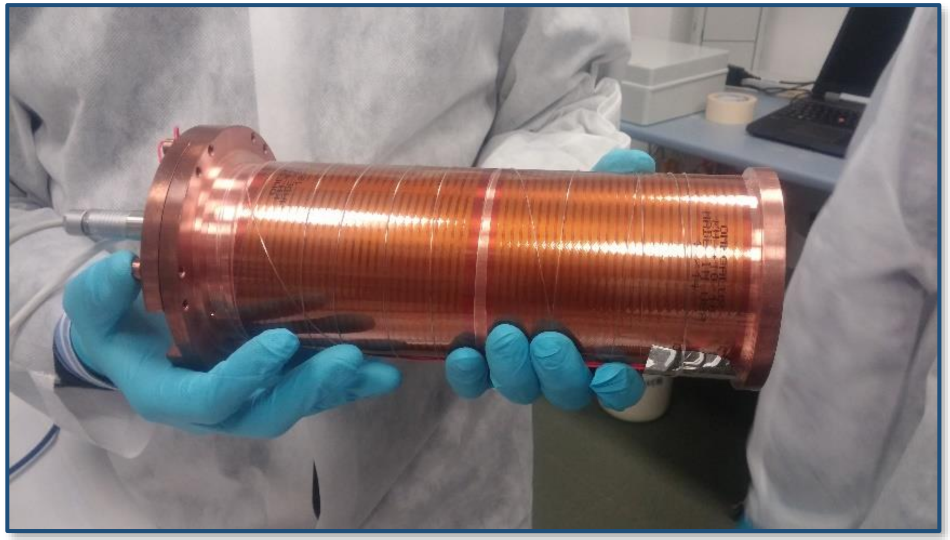


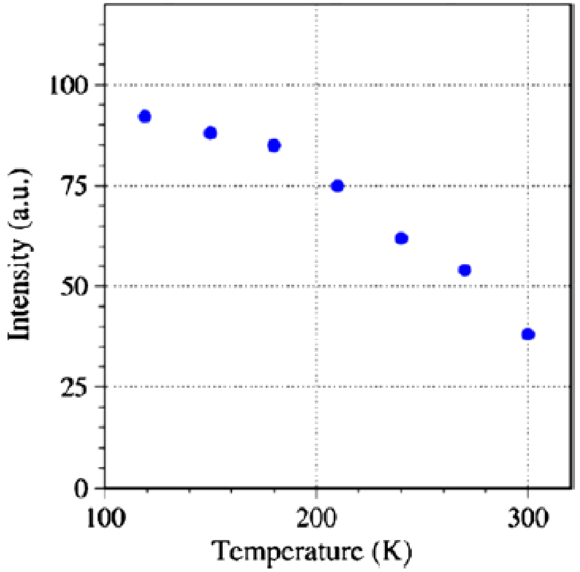
[32] Astrop. Phys. 33(2010)40

The anisotropy is significantly evident for oxygen nuclear recoils in the energy region down to hundreds keV at 5.4 σ confidence level

$ZnWO_4$ – 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)





Temperature dependence of the light output of the $ZnWO_4$ crystal scintillator for excitation with ^{241}Am α particles

NIM A **600** (2009) 594

- 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

- $ZnWO_4$ crystal scintillator is a promising detector for rare events search
- $ZnWO_4$ is one of the most radio-pure crystal scintillator
- The anisotropic features of the response of $ZnWO_4$ detectors can be exploited to investigate the directionality for DM candidates inducing nuclear recoils
- First evidence of anisotropy in the response of $ZnWO_4$ crystal scintillator for oxygen nuclear recoils in the energy region down to some hundreds keV measured at 5.4 σ confidence level
- These detectors can be used to obtain, with a completely different new approach, further evidence for the presence of DM candidates inducing nuclear recoils in the galactic halo and provide complementary information on the nature and interaction type of the DM candidate
- R&D and new measurements are in progress in the framework of the ADAMO project