Development of high-quality $ZnWO_4$ scintillating detectors to search for dark matter and double-beta decay

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Motivation

$ZnWO_4$ scintillators:

- “source=detector” geometry: detection efficiency up to 100% for internal α,β particles and nuclear recoils;
- relatively high scintillation efficiency;
- high radiopurity level;
- discrimination of α, nuclear recoils and β(γ) signals by using the scintillation signal shape;
- high anisotropy relative to heavy particles directionality (α, nuclear recoils).

Processes to investigate:

- DM directionality approach (ADAMO project [1])
- Study of α decay of W naturally occurring nuclides ($^{180,182,183,184,186}W$)
- Search for double-beta processes in $^{64,70}Zn$, $^{180,186}W$

[1] F. Cappella et al., Eur. Phys. J. C 73, 2276 (2013); P. Belli et al., Int. J. Mod. Phys. A 37(7), 2240013 (2022).

$ZnWO_4$ crystals production

$ZnWO_4$ crystals are grown by the low-thermal gradient Czochralski technique in the Nikolaev Institute of Inorganic Chemistry (NIIC, Novosibirsk, see [1] for details).

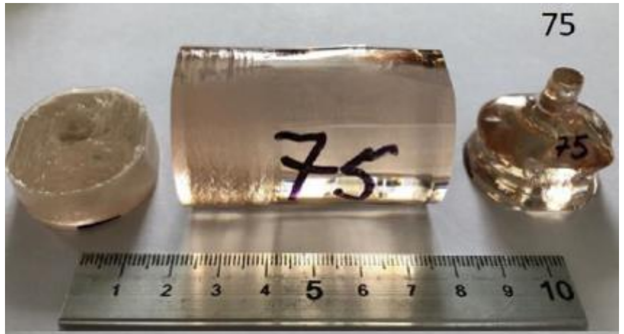
Different WO_3 compounds were used:

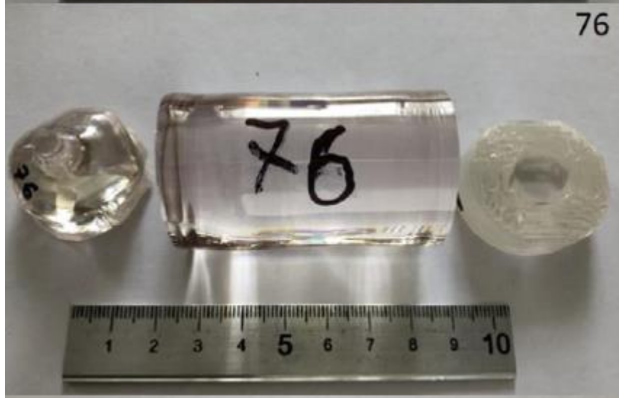
- NIIC I: <50 ppm of Si, ≤1 ppm of transition metals;
- NNIC II: additionally purified by tungsten chlorides sublimation;
- NT: manufactured by Nippon Tungsten Co., Ltd. (Japan);
- JNM: manufactured by Japan New Metals Co., Ltd., <1 ppm of Fe, <10 ppm of Mo

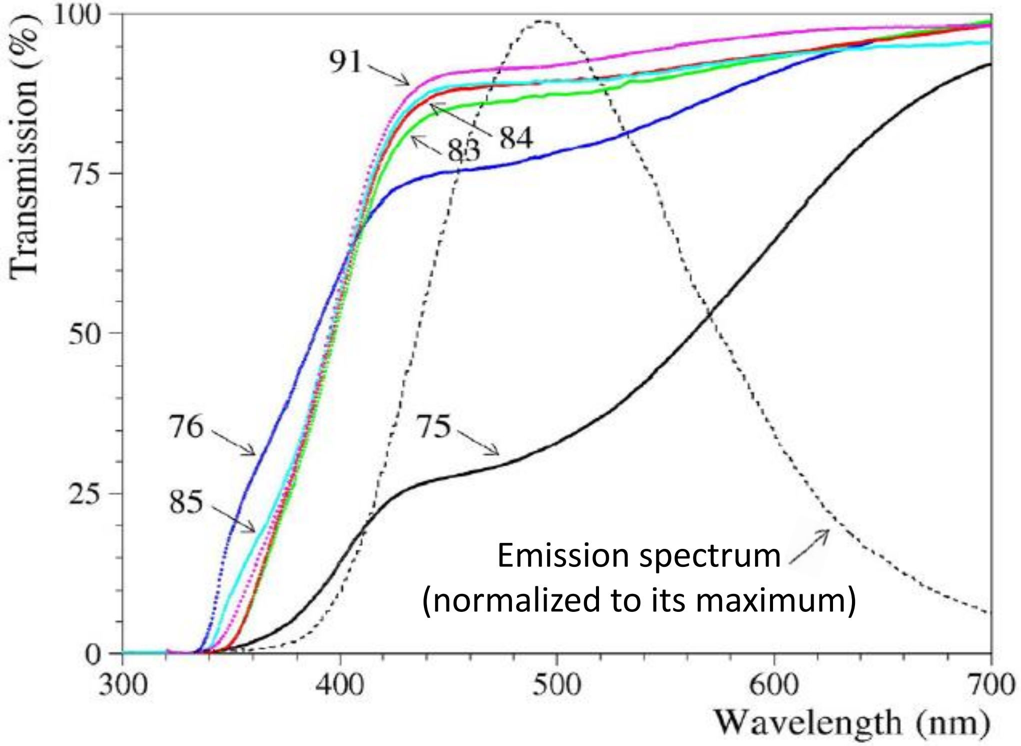
Crystal boule No.	**Sample size (mm^3)**	**Number of crystallisations**	**WO_3**	**Compound stoichiometry**
75	10×10×2 ⌀30×60	2	NIIC II	+0.3% of WO_3
76	10×10×2 ⌀30×60	2	NT	+0.25% of ZnO
83	10×10×2 ⌀30×60	1, annealed	NIIC I	+0.15% of WO_3
84	10×10×2 ⌀30×60	1, annealed	NIIC I	Stoichiometric
85	10×10×2 ⌀30×60	1, annealed	JNM	Stoichiometric
91	⌀30×67	1, annealed	NIIC I	Stoichiometric
94	⌀30×31 ⌀30×32	1, annealed	NIIC I	Stoichiometric

[1] P.Belli et al., Optical, luminescence, and scintillation properties of advanced $ZnWO_4$ crystal scintillators, Nucl. Instrum. Methods A 1029, 166400 (2022).

Optical transmission





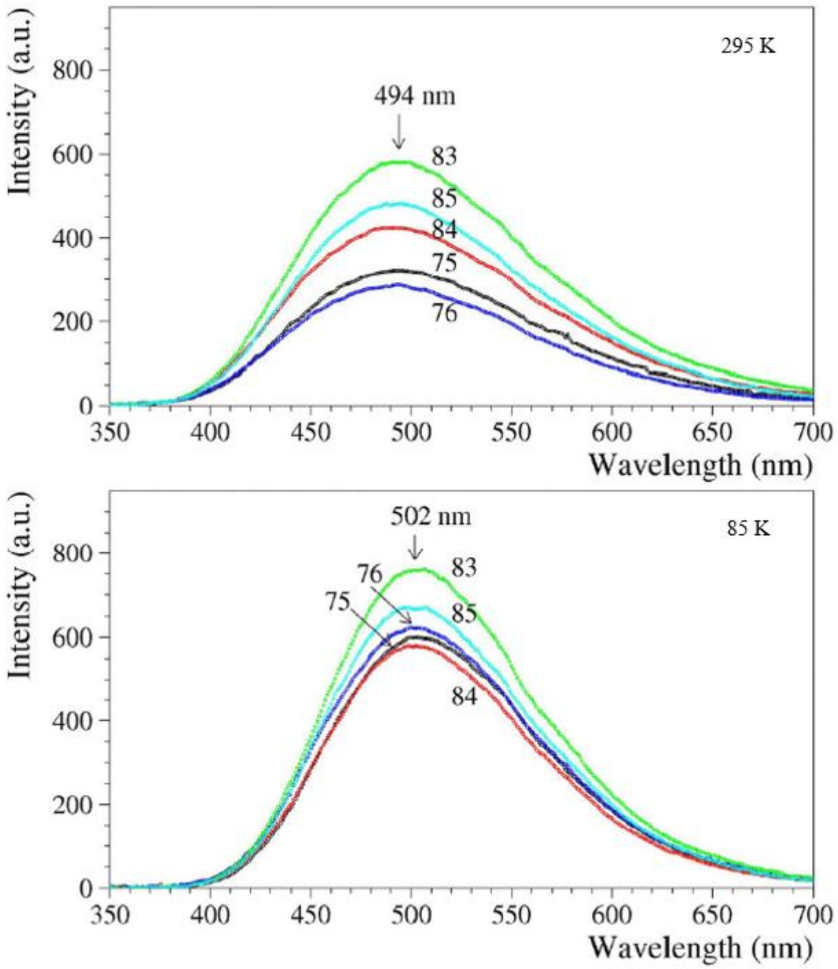


Emission spectrum
(normalized to its maximum)

- Boules No. 75, 76, 83-85: Shimadzu UV-2201 Double-beam UV spectrometer
- Boule No. 91: Perkin Elmer UV/VIS spectrometer Lambda 18
- Thin (1.0 to 1.8 mm) $ZnWO_4$ samples were used in the reference beams to correct light losses caused by the Fresnel reflection.

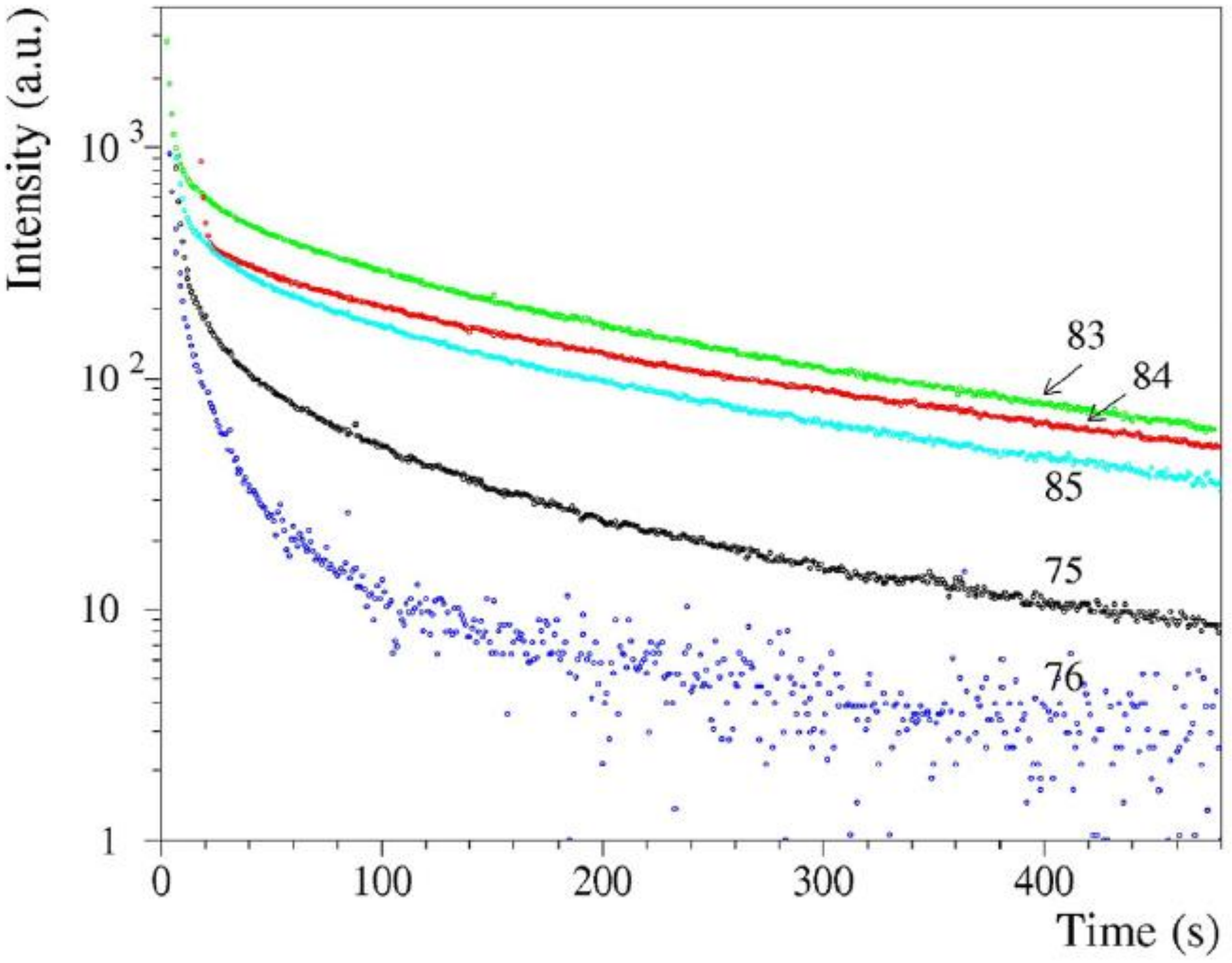
X-Ray luminescence (XRL)

- 10×10×2 mm^3 samples on the copper holder in the vacuum cryostat;
- X-ray tube with Cu anode
  - Operating voltage: 20 kV
  - Current: 25 mA
  - Flux: 0.25 mW/cm^2
- Temperature control: chromel-copel thermocouple, semiconductor silicon sensor.
- Luminescence detection:
  - PMT: FEU-106 (sensitive in the region 350-820 nm)
  - Monochromator MDR-2 (600 mm^{-1} diffraction grating)



Emission spectra of the samples under X-ray irradiation

Phosphorescence

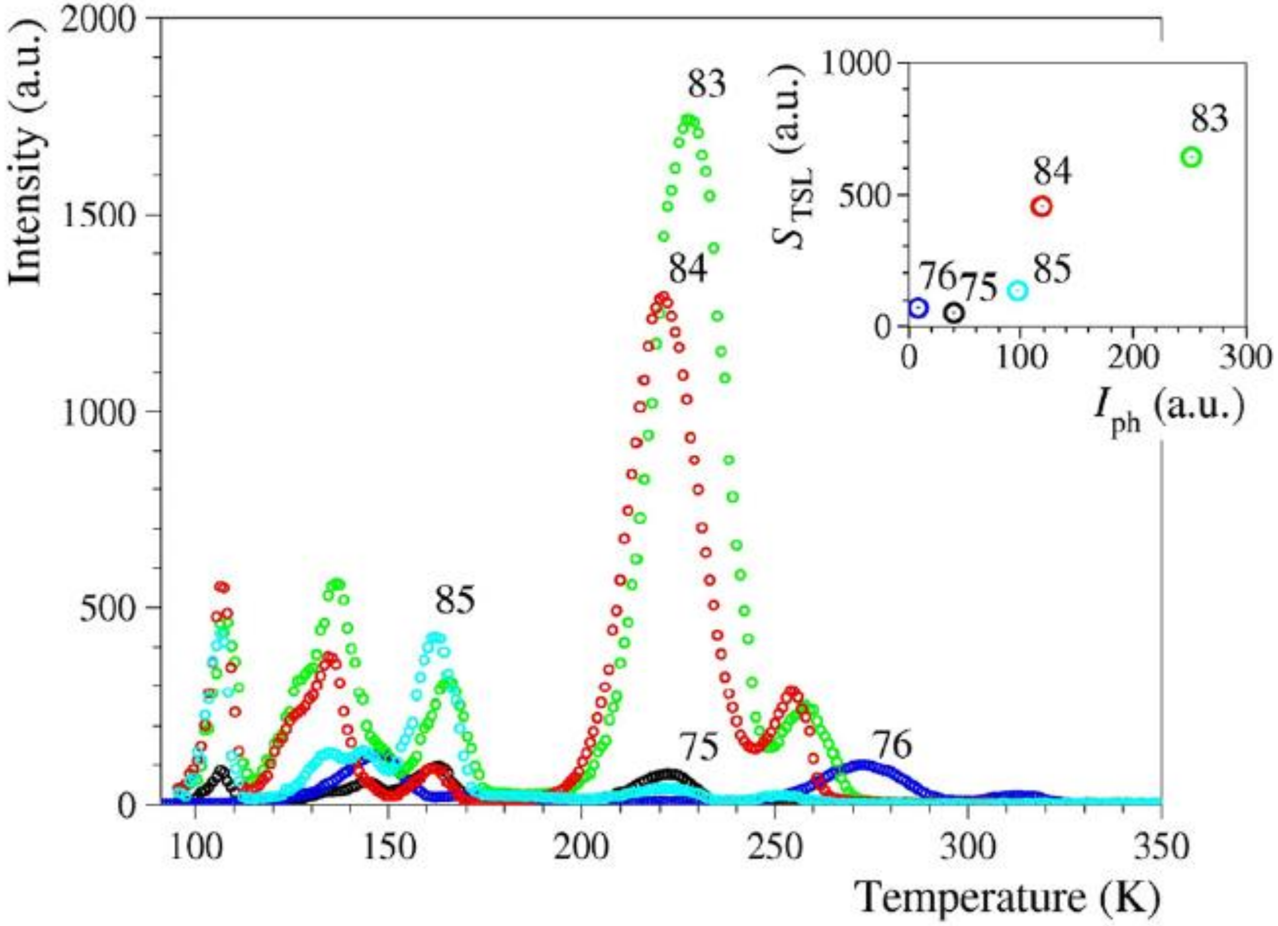


XRL phosphorescence of the $ZnWO_4$ samples measured at 85 K after X-ray irradiation over 20 min

- the sensitivity of the recording system to register the phosphorescence was increased by two orders of magnitude → → the concentration of the traps that cause phosphorescence are orders of magnitude lower than that of the radiative recombination centers in the $ZnWO_4$ crystals.
- No detectable phosphorescence after irradiation at 295 K.

Thermally stimulated luminescence (TSL)

- X-ray irradiation: 20 min (0.30 J/cm^2), 85 K; heating rate: (0.30 ± 0.02) K/s.
- All the samples have a similar set of traps, however with different concentration (much lower in the samples 75-76 grown by re-crystallisation).
- Some correlation between the TSL and phosphorescence intensities indicates that the concentration of all kinds of traps in the crystal varies depending on the growing technology.
- No detectable TSL after irradiation at 295 K.

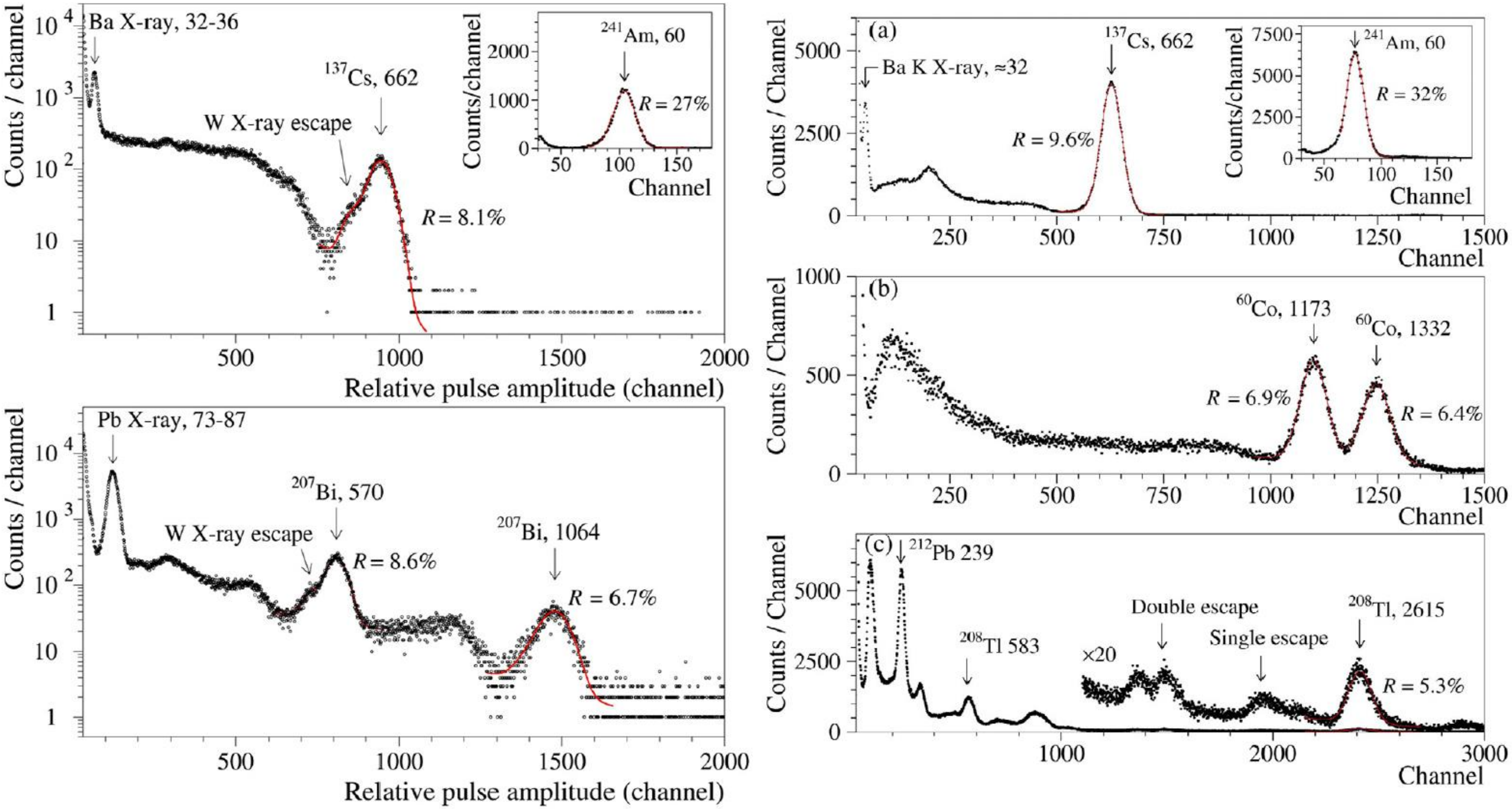


Thermally stimulated luminescence of $ZnWO_4$ samples measured after X-ray irradiation. Inset: area of TSL curves in the temperature interval 95–320 K (S_{TSL}) versus phosphorescence intensity at 200 s after the X-ray irradiation termination (I_{ph}).

Scintillation properties

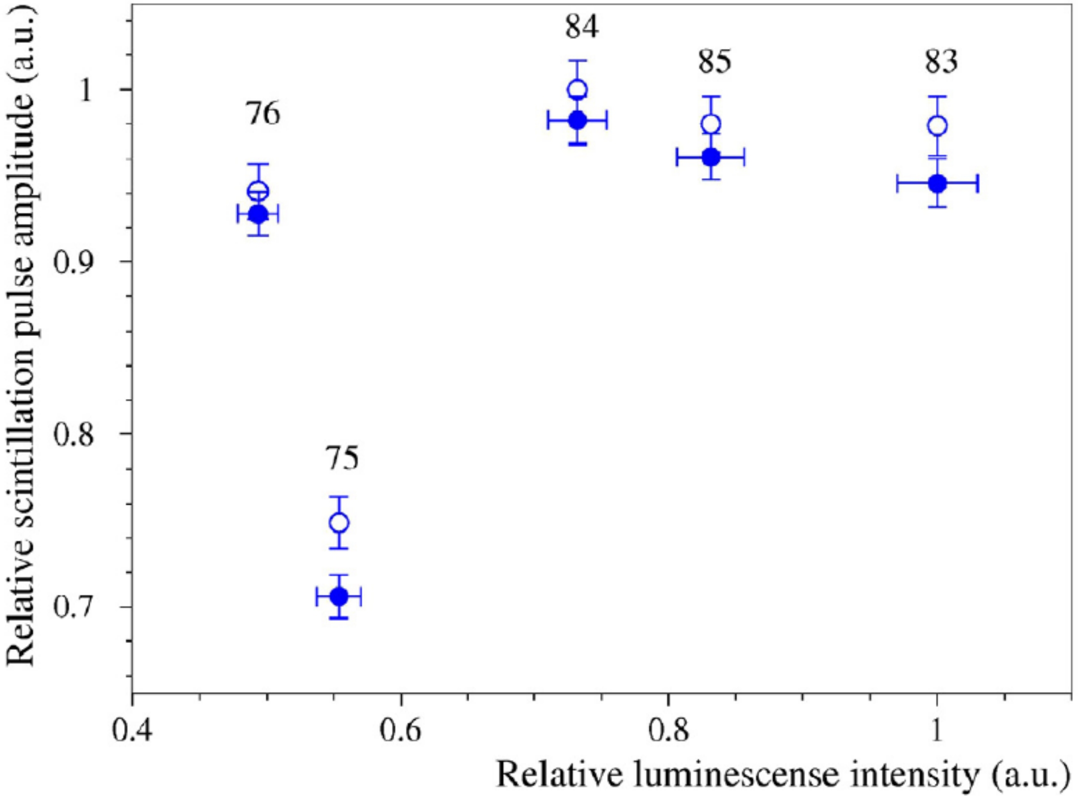
- 3″ PMT Hamamatsu R6233-100: bialkali photocatode, spectral response 300-650 nm (maximum at 420 nm).

- Spectroscopy amplifier (15-μs shaping time), peak sensitive analog-to-digital converter (both produced by the Intelligent Electronic Systems, Kharkiv).



Scintillation spectra measured with the small sample No. 84 (10×10×2 mm^3, left panel) and the large sample No. 94 (⌀30×31 mm^3, right panel) by using different γ-ray sources.

Scintillation vs. XRL



Relative scintillation pulse amplitude versus relative XRL intensity of the $ZnWO_4$ crystal samples at room temperature (filled circles). Results of scintillation measurements with non-irradiated samples are shown by open circles.

- Relative scintillation and XRL intensities are not correlated. It could be explained by the contribution of phosphorescence (does not contribute to the scintillation pulse amplitude), a dose dependence of XRL intensity (negligible impact in scintillation measurements).
- Scintillation response of the samples irradiated by X-rays is (1.4-5.7)% lower.

Conclusion

- Optical, luminescence and scintillation properties have been measured with several $ZnWO_4$ crystal scintillators grown by the low-thermal gradient Czochralski technique after an extended R&D, including variations of the compound stoichiometry, using of WO_3 of different producers, utilization of re-crystallization and annealing of the grown boules.
- The best optical and scintillation properties have been obtained with the crystal samples produced by single crystallization with the stoichiometric composition of the $ZnWO_4$ compound prepared from deeply purified WO_3, annealed in air atmosphere.
- No clear correlation was observed between the scintillation and luminescence relative intensities, which indicates that the material quality can be improved further. The energy resolution (full weight at half maximum, FWHM) has been measured with a $\varnothing 30\times 31$ mm sample as 9.6% for 661.7 keV γ quanta of ^{137}Cs, and 6.4% at 1332.5 keV (^{60}Co). The measurements with two highest quality crystal samples are in progress at the Gran Sasso underground laboratory to estimate radiopurity level of the samples.
- Measurements details and theoretical explanations of luminescence mechanisms have been published in [1, 2].

[1] P.Belli et al., Optical, luminescence, and scintillation properties of advanced $ZnWO_4$ crystal scintillators, Nucl. Instrum. Methods A 1029 (2022) 166400.

[2] V. Ya. Degoda, Luminescence of $ZnWO_4$ crystals under X-ray excitation, J. Lumin. 249 (2022) 119028.