Search for inelastic Dark Matter induced deexcitations of ^{178m2}Hf isomer in the Gran Sasso underground laboratory

P. Belli[a,b], R. Bernabei[a,b], F. Cappella[c,d], V. Caracciolo[a,b], R. Cerulli[a,b], F.A. Danevich[a,e], S.S. Drapey[e], A. Incicchitti[c,d], V.I. Kyryshchuk[e], S.I. Kyryshchuk[f] , M. Laubenstein[g], A. Leoncini[a,b], V.I. Tretyak[e,g]

[a] *INFN sezione Roma "Tor Vergata", Rome, Italy*

[b] *Dipartimento di Fisica, Universita di Roma "Tor Vergata', Rome, Italy*

[c] *INFN sezione Roma, Rome, Italy*

[d] *Dipartimento di Fisica, Universita di Roma "La Sapienza", Rome, Italy*

[e] *Institute for Nuclear Research of NASU, Kyiv, Ukraine*

[f] *Johannes Gutenberg University, Mainz, Germany*

[g] *INFN, Laboratori Nazionali del Gran Sasso, Assergi (AQ), Italy*

MEDEX'2025, Prague, Czech Republic, 23-27.06.2025

Idea to search for the collisional deexcitation of metastable nuclear isomers by the inelastic Dark Matter (iDM) particles.
Suggested isomers:
^{180m}Ta ("stable", $T_{1/2}$>2.9×10^{17} yr, nat. abundance 0.012%)
^{178m2}Hf (31 yr),
^{177m}Lu (160 d),
^{137m}Ba (2.5 min).

Experimental searches for possible DM induced decay of ^{180m}Ta

PHYSICAL REVIEW D **101,** 055001 (2020)

Editors' Suggestion | Featured in Physics

Metastable nuclear isomers as dark matter accelerators

Maxim Pospelov,[1,2] Surjeet Rajendran,[3] and Harikrishnan Ramani[4,5,*]

PHYSICAL REVIEW LETTERS **124,** 181802 (2020)

Search for Dark Matter Induced Deexcitation of ^{180}Ta^{m}

Björn Lehnert,[1,*] Harikrishnan Ramani,[2,3,†] Mikael Hult,[4] Guillaume Lutter,[4] Maxim Pospelov,[5,6,7] Surjeet Rajendran,[8] and Kai Zuber[9]

PHYSICAL REVIEW LETTERS **131,** 152501 (2023)

Constraints on the Decay of ^{180m}Ta

I. J. Arnquist,[1] F. T. Avignone III,[2,3] A. S. Barabash,[4] C. J. Barton,[5] K. H. Bhimani,[6,7] E. Blalock,[8,7] B. Bos,[6,7] M. Busch,[9,7] M. Buuck,[10] T. S. Caldwell,[6,7] C. D. Christofferson,[11] P.-H. Chu,[12] M. L. Clark,[6,7] C. Cuesta,[13] J. A. Detwiler,[10] Yu. Efremenko,[14,3] H. Ejiri,[15] S. R. Elliott,[12] G. K. Giovanetti,[16] J. Goett,[12] M. P. Green,[8,7,3] J. Gruszko,[6,7] I. S. Guinn,[6,7] V. E. Guiseppe,[3] C. R. Haufe,[6,7] R. Henning,[6,7] D. Hervas Aguilar,[6,7] E. W. Hoppe,[1] A. Hostiuc,[10] I. Kim,[12,*] R. T. Kouzes,[1] T. E. Lannen V.,[2] A. Li,[6,7] J. M. López-Castaño,[3] R. Massarczyk,[12,7] S. J. Meijer,[12] W. Meijer,[12] T. K. Oli,[5,2] L. S. Paudel,[5] W. Pettus,[17] A. W. P. Poon,[18] D. C. Radford,[3] A. L. Reine,[6,7] K. Rielage,[12] A. Rouyer,[16] N. W. Ruof,[10,*] D. C. Schaper,[12] S. J. Schleich,[11] T. A. Smith-Gandy,[16] D. Tedeschi,[2] J. D. Thompson,[11] R. L. Varner,[3] S. Vasilyev,[19] S. L. Watkins,[12] J. F. Wilkerson,[6,7,3] C. Wiseman,[10] W. Xu,[5] and C.-H. Yu[3]

(MAJORANA COLLABORATION)

D. S. M. Alves,[12] L. Hebenstiel,[12,17] and H. Ramani[20]

Eur. Phys. J. C (2023) 83:925
https://doi.org/10.1140/epjc/s10052-023-12087-5

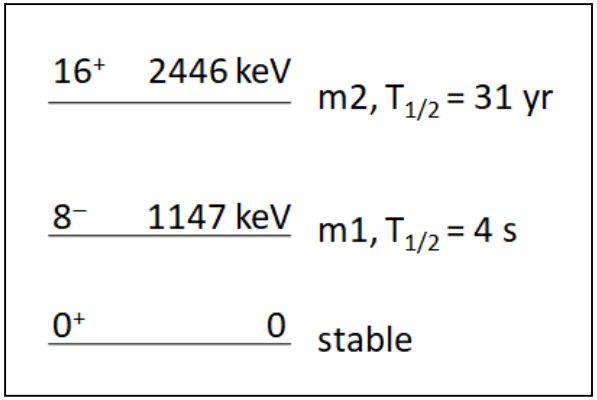
THE EUROPEAN PHYSICAL JOURNAL C

Regular Article - Experimental Physics

Deep-underground search for the decay of ^{180m}Ta with an ultra-low-background HPGe detector

R. Cerroni[1], S. Dell'Oro[2,3], A. Formicola[4], S. Ghislandi[1,5], L. Ioannucci[6], M. Laubenstein[1,a], B. Lehnert[7], S. S. Nagorny[8,9], S. Nisi[1], L. Pagnanini[1,5,9]

^{178m2}Hf is extremely interesting nucleus:
big energy (2446 keV) and big half-life (31 yr) of the 2nd excited level (because of high spin 16^+ and low energy of the 1st step in its decay)



Huge energy stored:
1 g of ^{178m2}Hf is equivalent to 300 kg of TNT explosive.
Ideas about possible military use ("hafnium bomb", 1990's).
Some physicists obtained good money for investigations …


APS NEWS

June 2007 (Volume 16, Number 6)

The Back Page

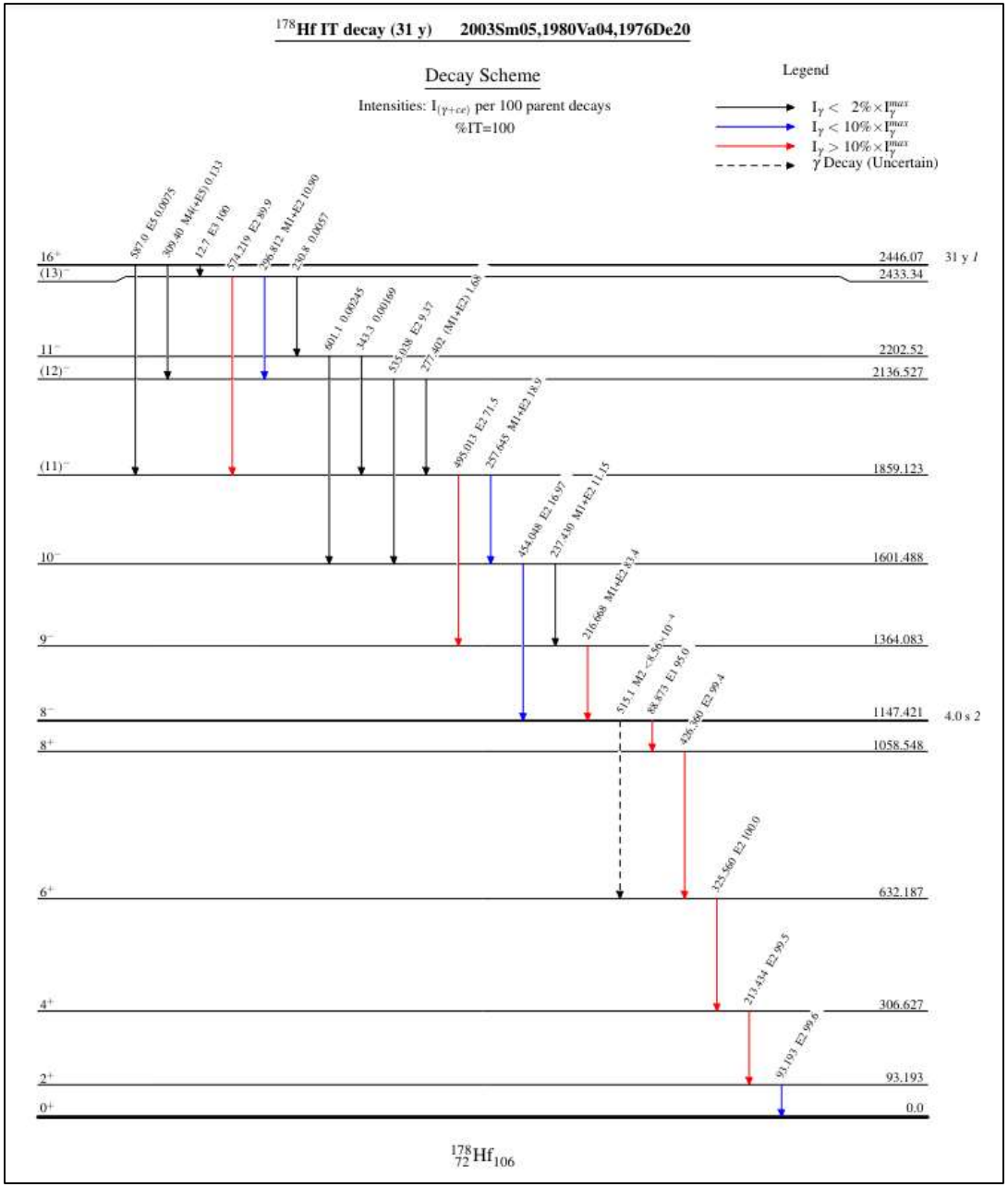
The Strange Tale of the Hafnium Bomb: A Personal Narrative

By Peter D. Zimmerman

Attempts to trigger its decay (by α's, e's, γ's, …).
Controversary results: claims for observing some enhancement (~4%, e.g. [1] and no observations of the effect (see e.g. review [2]).

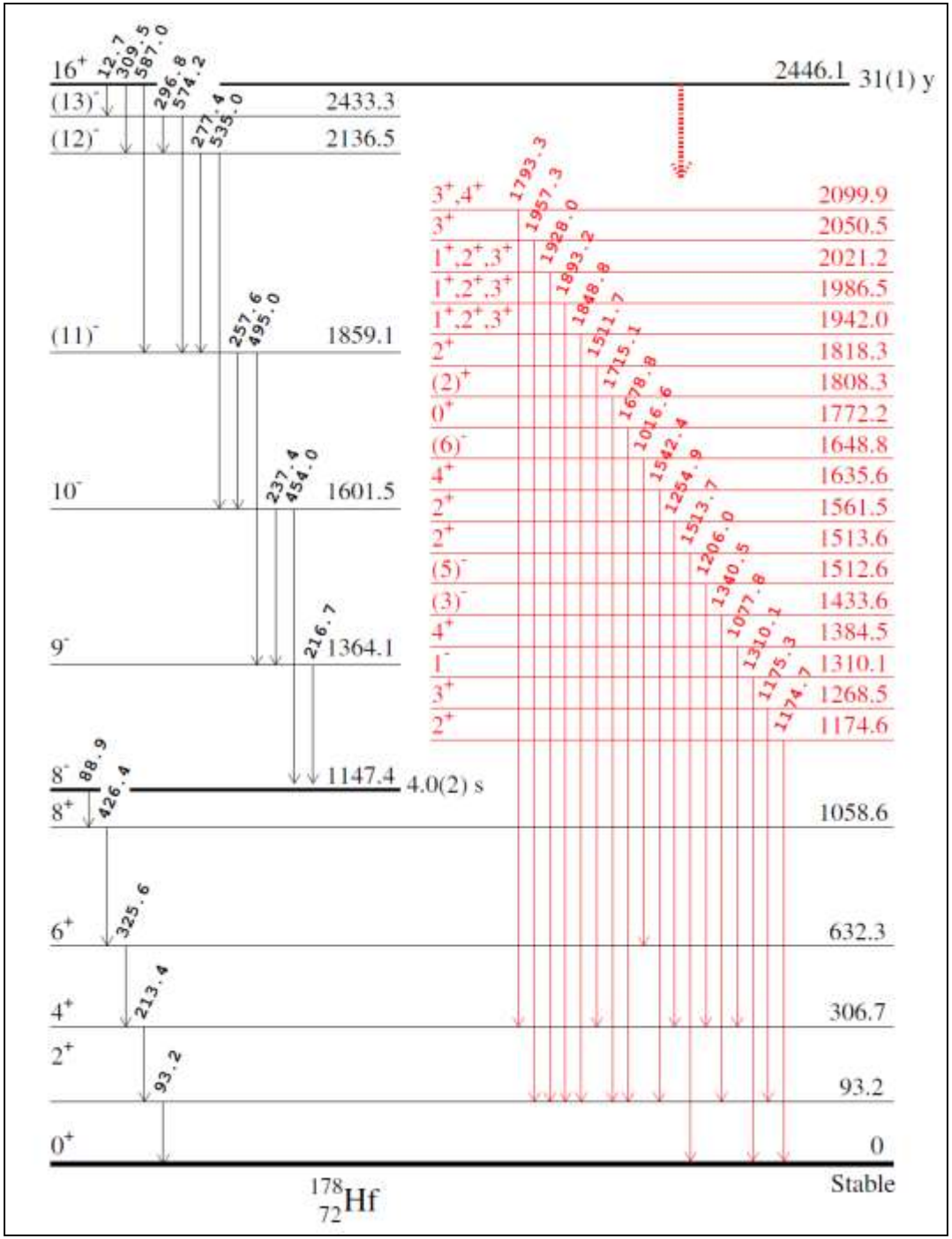
1. C.B. Collins et al., Accelerated Emission of Gamma Rays from the 31-yr Isomer of ^{178}Hf Induced by X-Ray Irradiation, Phys. Rev. Lett. 82 (1999) 695)
2. J.J. Carroll, An experimental perspective on triggered gamma emission from nuclear isomers, Laser Phys. Lett. 1 (2004) 275.

Usually ^{178m2}Hf decays by IT with emission of cascade of γ's, the most energetic has E_γ = 601.1 keV [1]

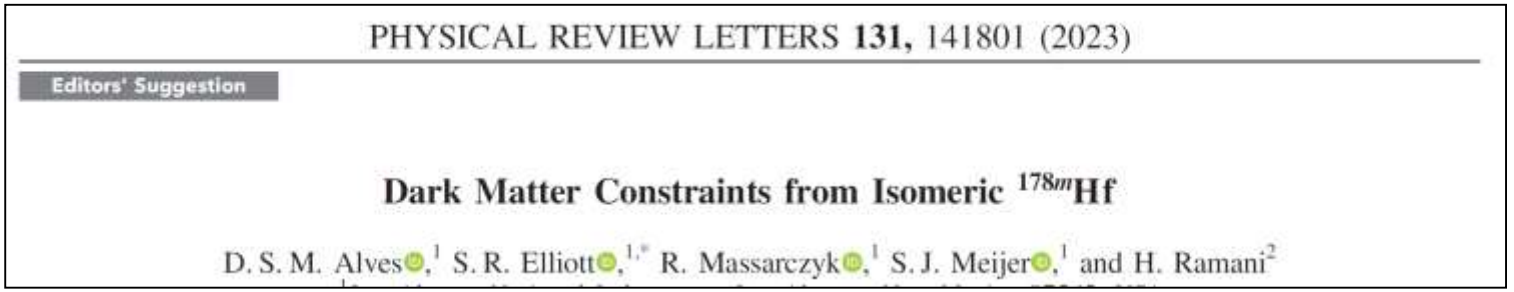


1. E. Achterberg, O.A. Capurro, G.V. Marti , Nucl. Data Sheets 110 (2009) 1473.

Possible interactions of ^{178m2}Hf with some Dark Matter candidates (iDM, SIDM, …) could lead to emission of γ's with higher energies (>1 MeV) from the excited levels of ^{178}Hf which are not populated in its usual IT decay.



First experimental search for DM induced decay of ^{178m2}Hf



PHYSICAL REVIEW LETTERS **131,** 141801 (2023)

Editors' Suggestion

Dark Matter Constraints from Isomeric ^{178m}Hf

D. S. M. Alves,[1] S. R. Elliott,[1,*] R. Massarczyk,[1] S. J. Meijer,[1] and H. Ramani[2]

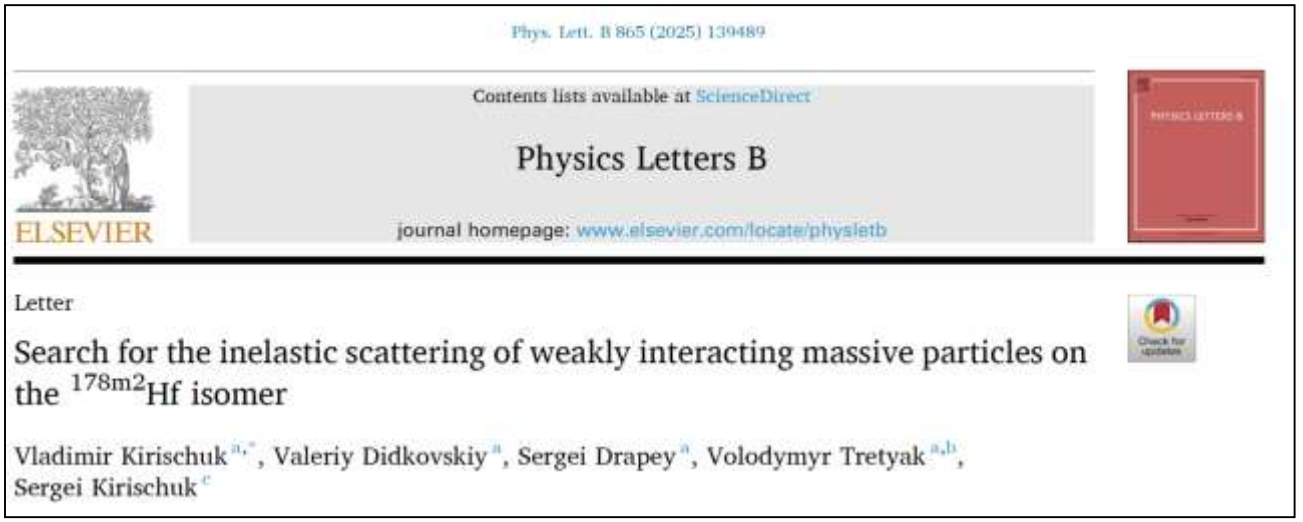
LANL, 1 kg of natural Ta was irradiated by p beam during 60 days (1440 h) in 1981,
E_p = ~800 MeV, chemical separation of Hf [1]
~4×10^7 Bq of ^{178m2}Hf (1.7×10^{-7} g)

^{178m2}Hf source is still very hot, so 1.2 m distance between HPGe detector,
20 min of measurements on the Earth's surface level

Search for peaks at 11 characteristic energies > 1 MeV (deexcitation of ^{178m2}Hf levels not populated in usual IT decay)
$T_{1/2}$ limits > $(0.1–1.8)\times10^5$ yr

1. W.A. Taylor et al., Recovery of hafnium radioisotopes from a proton irradiated tantalum target, J. Radioanalytical and Nucl. Chem. 236 (1998) 155.

These $T_{1/2}$ limits were improved very recently by ~1 order of magnitude in INR NASU experiment



Phys. Lett. B 865 (2025) 139489

Contents lists available at ScienceDirect

Physics Letters B

journal homepage: www.elsevier.com/locate/physletb

Letter

Search for the inelastic scattering of weakly interacting massive particles on the ^{178m2}Hf isomer

Vladimir Kirischuk [a,*], Valeriy Didkovskiy [a], Sergei Drapey [a], Volodymyr Tretyak [a,b], Sergei Kirischuk [c]

Institute for Nuclear Research of the Nat. Ac. Sci. of Ukraine

^{178m2}Hf source was created by irradiation of Ta foil 300 μm thick by 1.2 GeV electron beam at the Kharkiv linear accelerator in 1966 – 1970 (10,000 h of irradiation)

Current activity ~70 Bq

BEGe detector ~70 cm^3, 10^6 s, Earth's surface level

11 excited levels from the previous work + 6 new, no effect, $T_{1/2}$ limits > $(2.3–9.2)\times10^5$ yr

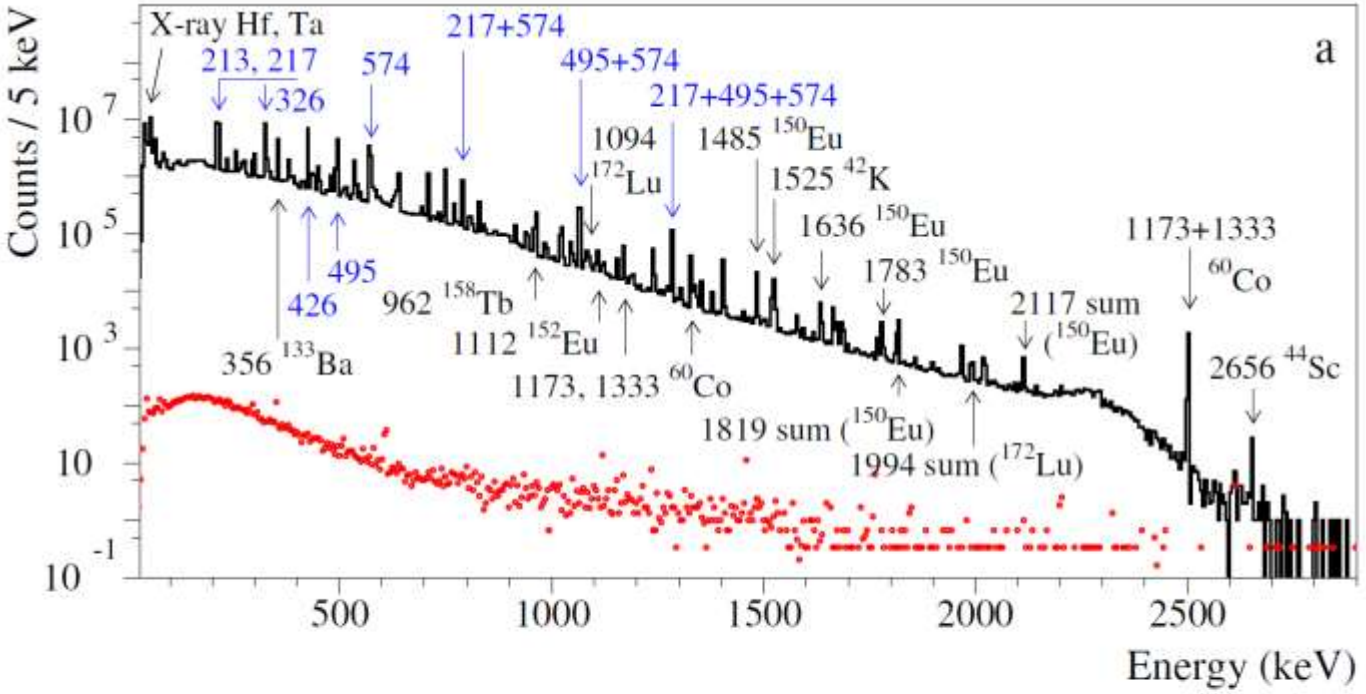
Current work: first underground measurement of ^{178m2}Hf source in low-background conditions → lower background at > 1 MeV, higher sensitivity

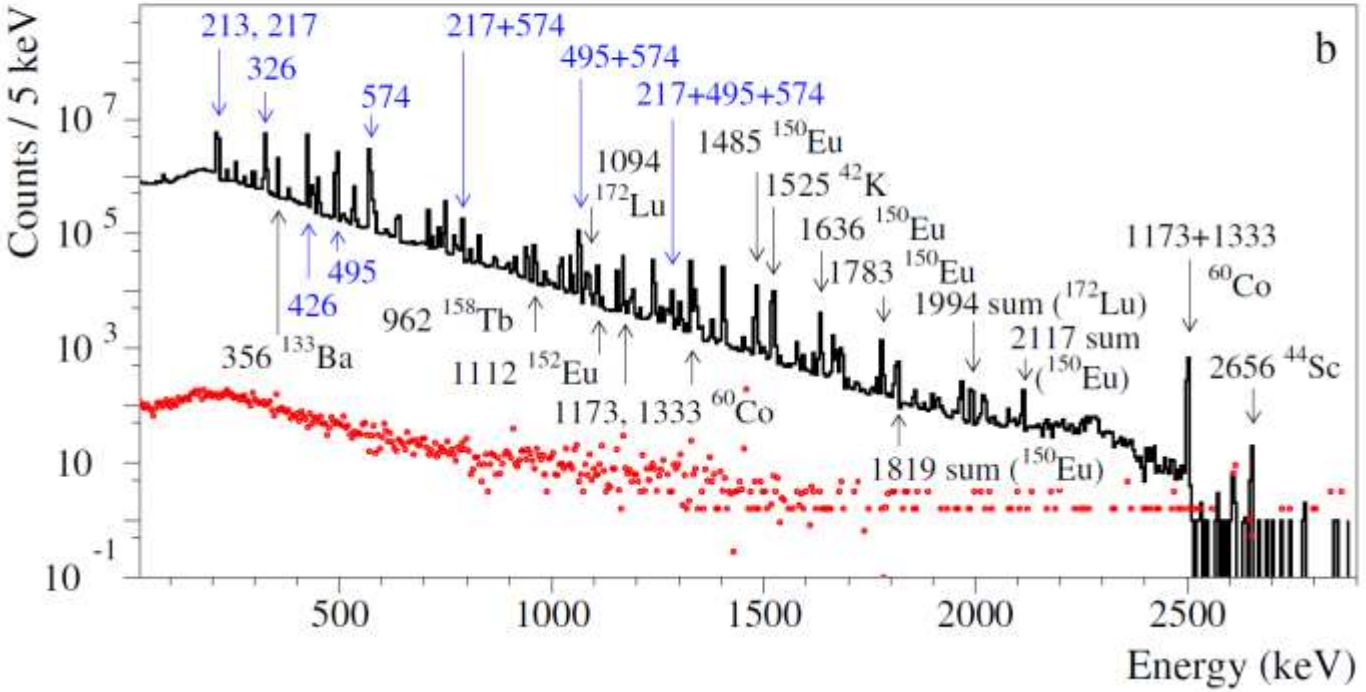
Two measurements at LNGS (3800 m w.e.) with INR NASU source

(1) BEGe detector 118 cm^3, shielding: HP Cu 5 cm, Pb 20 cm, Plexiglas box flushed with pure N_2, 558 h ("zero" geometry)

Better sensitivity to low energy region to look for some other interesting processes; many sum peaks and X rays

(2) HPGe GeCris 465 cm^3, shielding: HP Cu 5 cm, Pb 25 cm, Plexiglas box flushed with pure N_2, 612 h
To suppress summations effects: 22.6 mm distance
To suppress X rays: 2.4 mm Cu plates (+1.5 mm Cu endcap)
Better efficiency in higher energy region





(a) BEGe 558 h
(b) GeCris 612 h

Peaks from ^{178m2}Hf – in blue

In red – backgrounds of BEGe and GeCris detectors

^{42}K (daughter of ^{42}Ar, $T_{1/2}$ = 33 yr),
^{44}Sc (^{44}Ti, 59 yr),
^{60}Co (5.3 yr),
^{133}Ba (10.5 yr),
^{150}Eu (37 yr),
^{152}Eu (13.5 yr),
^{158}Tb (180 yr),
^{172}Lu (^{172}Hf, 1.9 yr)
…
are clearly visible

> 100 peaks at > 1 MeV

Decryption of the spectrum:

(1) Correspondence of the measured E_γ to the table value;

(2) Correspondence of the peak area to the table intensity (if nuclide has a few peaks);

(3) Comparison of the measured activity and measurements in Kyiv in 2012 - $T_{1/2}$ expectation

Big work …

2 peaks are still unidentified:

- 1561.8(4) keV – 75(19) counts, $T_{1/2}$ ~ 5.8(19) yr
- 1567.5(4) keV – 39(18) counts, $T_{1/2}$ ~ 4.1(14) yr

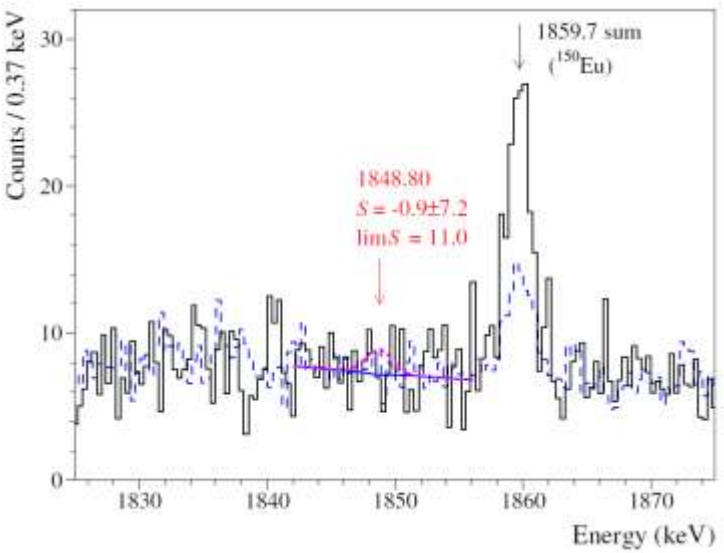
Radionuclide	Decay modes, Branching ratio (%)	Half-life (yr)	Activity (Bq)
^{178m2}Hf	IT 100	31(1)	62.6(12)
^{42}K (^{42}Ar)	β^- 100	32.9(11) (^{42}Ar)	1.47(8)
^{44}Sc (^{44}Ti)	EC 100	59.1(3) (^{44}Ti)	0.383(20)
^{60}Co	β^- 100	5.2712(4)	0.620(25)
^{101}Rh	EC 100	3.3(3)	0.034(8)
^{102m1}Rh	EC β^+ 99.767(24) IT 0.233(24)	3.742(10)	0.090(8)
^{108m}Ag	EC β^+ 91.3(9) IT 8.7(9)	438(9)	0.544(17)
^{133}Ba	EC 100	10.551(11)	22.8(7)
^{150}Eu	EC β^+ 100	36.9(9)	11.12(34)
^{152}Eu	EC β^+ 72.08(13) β^- 27.92(13)	13.517(9)	2.25(6)
^{154}Eu	β^- 99.982(12) EC β^+ 0.018(12)	8.601(10)	0.197(7)
^{158}Tb	EC β^+ 83.4(7) β^- 16.6(7)	180(11)	2.15(7)
^{172}Lu (^{172}Hf)	EC 100	1.87(3)	0.418(23)
^{173}Lu	EC 100	1.37(1)	≤ 1.8
^{174}Lu	EC β^+ 100	3.31(5)	1.29(7)

Half-life limits on the iDM or SIDM induced transitions

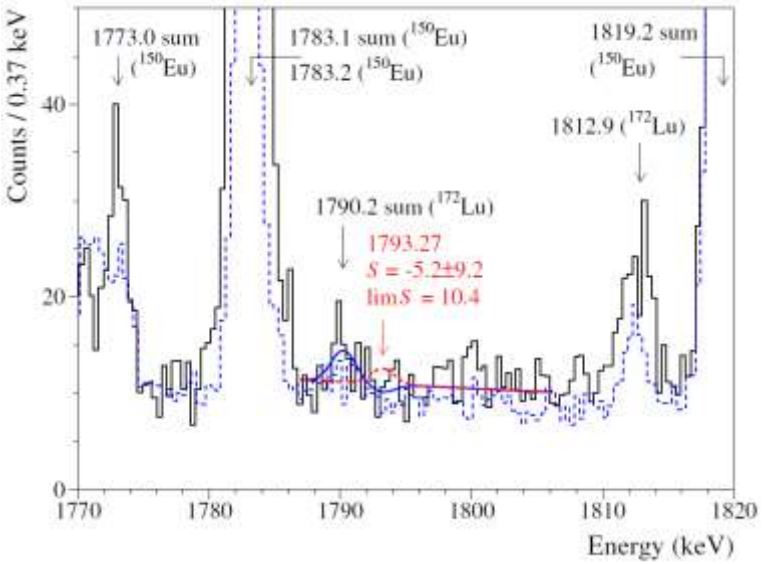
In general, we do not see peaks which could be ascribed to transitions induced by iDM and/or SIDM

$\lim T_{1/2} = T_{1/2}(^{178m2}Hf) \times (S_{495}/\lim S_\gamma) \times (b_\gamma/b_{495}) \times (\varepsilon_\gamma/\varepsilon_{495})$

$T_{1/2}(^{178m2}Hf) = 31$ yr
$S_{495} = 4.0\times10^6$ counts
b – branching ratio for the 495 keV peak or for the given γ
ε – efficiency for the 495 keV peak or for the given γ

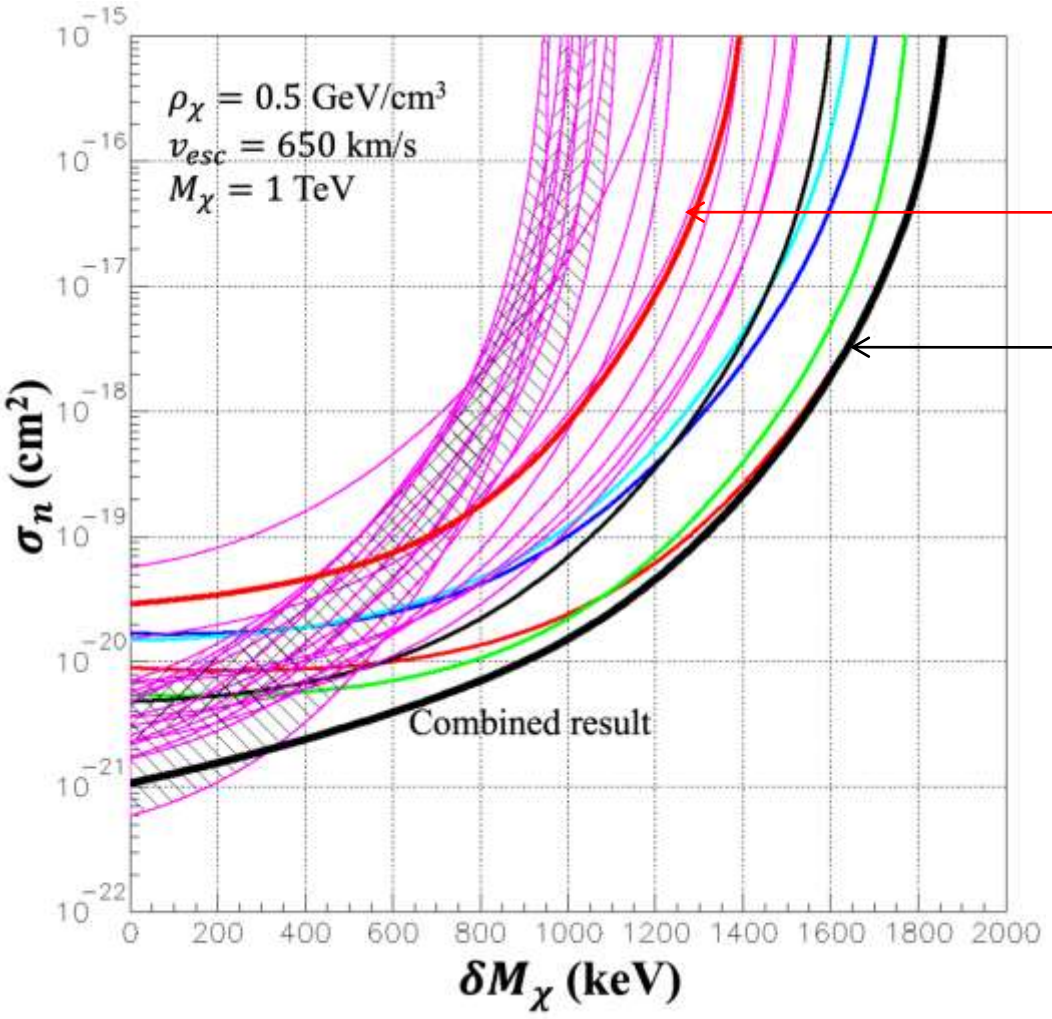


GeCris 612 h
(BEGe 558 h / 5 by blue dashed line)
$1^+,2^+,3^+$ 1942.007 keV $\rightarrow$ 2^+ 93.1803 keV
E_γ = 1848.80 keV
lim S = 11 counts, $T_{1/2} > 7.6\times10^6$ yr



GeCris 612 h
(BEGe 558 h / 5 by blue dashed line)
$3^+,4^+$ 2099.874 keV $\rightarrow$ 4^+ 306.6182 keV
E_γ = 1793.27 keV
lim S = 10.4 counts, $T_{1/2} > 15.8\times10^6$ yr

For 18 considered ^{178m2}Hf levels, $T_{1/2}$ limits in range of $4.7\times10^{6} - 1.6\times10^{7}$ yr
better than those obtained in [1] ($1.3\times10^{4} - 1.8\times10^{5}$ yr) and [2] ($(1.6 - 9.3)\times10^{5}$ yr)



Exclusion limits at 90% C.L. of the inelastic cross section on nucleon for $\chi + {}^{178m2}\mathrm{Hf} \rightarrow \chi^* + {}^{178j}\mathrm{Hf}$ process

1. D.S.M. Alves et al., Dark Matter Constraints from Isomeric ^{178m}Hf , Phys. Rev. Lett. 131 (2023) 141801.
2. V. Kirischuk et al., Search for the inelastic scattering of weakly interacting massive particles on the ^{178m2}Hf isomer, Phys. Lett. B 865 (2025) 139489.

Conclusions

First ultra-low-background measurements of ^{178m2}Hf source (300 μm Ta foil with ~70 Bq ^{178m2}Hf activity) were performed at the LNGS underground laboratory. Two well-shielded Ge detectors were used (BEGe 118 cm^3, 558 h and GeCris 465 cm^3, 612 h).

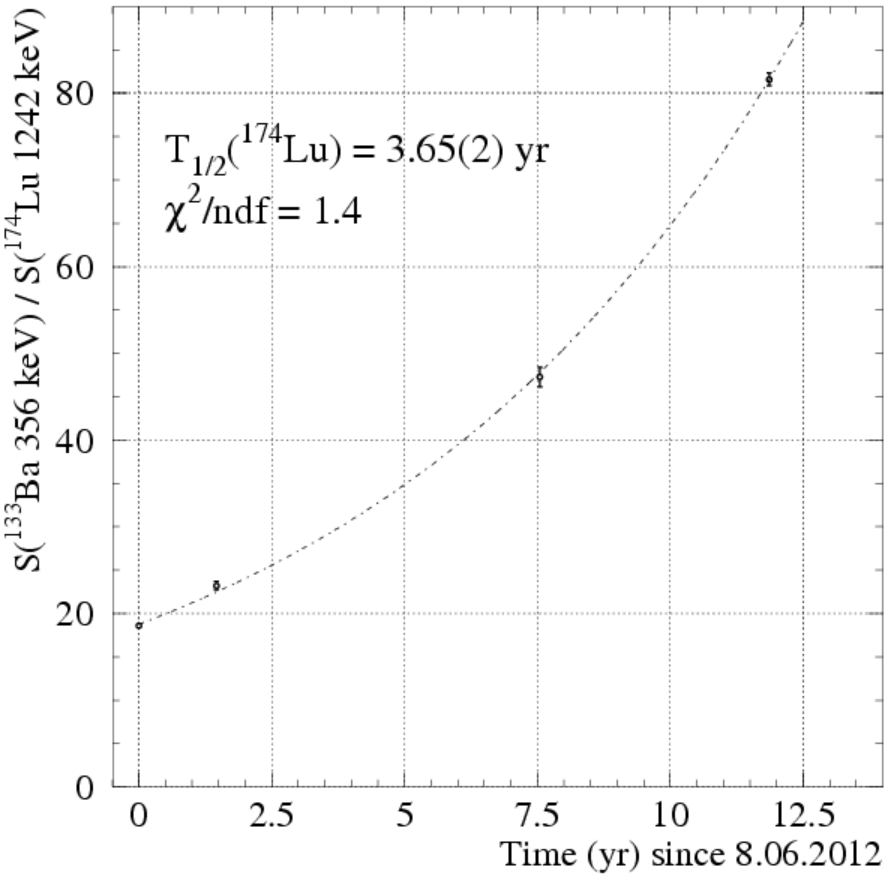
No effects were found which could be ascribed to transitions induced by possible interactions of ^{178m2}Hf with some DM candidates (iDM, SIDM).

Ultra-low-background conditions lead to higher sensitivity at > 1 MeV energies to possible iDM/SIDM induced transitions which lead to population (and subsequent deexcitation) of ^{178}Hf levels not involved in usual IT decay of ^{178m2}Hf. T1/2 limits for such transitions were set in range of 4.7×10^6 – 1.6×10^7 yr (preliminary), improved by 1 – 2 orders of magnitude comparing with the previous 2 experiments (done at the Earth's level and with not-specially-shielded detectors).

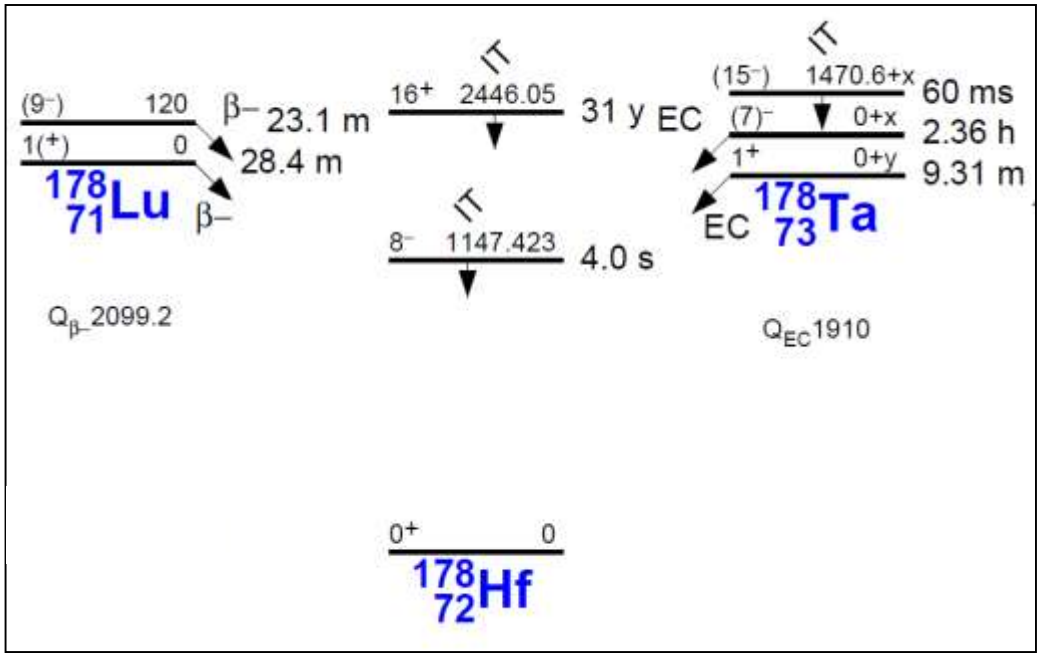
We have a plan to repeat such measurements with ^{178m2}Hf source, radiochemically extracted from Ta foils, which contains much lower amounts of other radioactive contaminations. The source is already available.

Some interesting additional results are also expected:
- search for rare decays of ^{178m2}Hf (β^-, EC, α)
- rare decays of some radioactive nuclides-contaminants present in the source
- more exact $T_{1/2}$ values for some nuclides (comparing with old INR NASU measurements)

Thank you for attention!
Děkuji za pozornost!



Current value of ^{174}Lu $T_{1/2}$ = 3.31(5) yr [NDS 87 (1999) 15] is based mainly on the value $T_{1/2}$ = 3.31(5) yr which in fact is not published (private communication of D. Nethaway 1973).



Energetically possible for ^{178m2}Hf are decays:

- IT (observed, well investigated)
- EC to ^{178}Lu (which subsequently decays to ^{178}Hf), $T_{1/2} > 3.1\times10^{3}$ yr [1]
- β^- to ^{178}Ta (with subsequent decay to ^{178}Hf), $T_{1/2} > 1.0\times10^{4}$ yr [1]
- α decay to ^{174}Yb (to excited levels due to spin difference), observed in [2] with $T_{1/2} = (2.5\pm0.5)\times10^{10}$ yr not in a good agreement with some theoretical calculations (~10^6 yr)

1. J. van Klinken et al., K-forbidden decays in ^{178}Hf; M4 decay of an yrast state, Nucl. Phys. A 339 (1980) 189.
2. S.A. Karamian et al., Weak K hindrance manifested in α decay of the ^{178}Hf^{m2} isomer, Phys. Rev. C 75 (2007) 057301.