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

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The 2^{nd} isomeric state of ^{178}Hf has high energy of 2.446 MeV, and big half-life of 31 yr related to its high spin 16^+. Normally it decays spontaneously to the ground state of ^{178}Hf by isomeric transition with emission of cascade of gamma quanta with energies up to $\sim$ 600 keV. Possible interactions of ^{178m2}Hf with some given inelastic Dark Matter (iDM) candidates could lead to emission of gamma rays with higher energies (>1 MeV) from the excited levels of ^{178}Hf which are not populated in its usual decay. The search for such gamma rays was performed deep underground at the Gran Sasso National Laboratory of the INFN in measurements of ^{178m2}Hf source with an activity of $\simeq$ 63 Bq with two ultra-low-background HPGe gamma spectrometers over cumulative 1170 h. This is the first ^{178m2}Hf measurements in the low-background conditions which allowed the enhancement of the sensitivity — in the framework of the models considered here — to hypothesized iDM induced decay at energies >600 keV thanks to reduced natural radioactive background. Improved $T_{1/2}$ limits for several hypothesized iDM induced transitions were set, up to the level of $\lim T_{1/2} \sim 10^7$ yr, higher (or competitive) than those known previously.

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PoS(MEDEX2025)037

1. Introduction

While the existence of Dark Matter (DM) is well established by the studies of gravitational interaction on different spatial scales and by anisotropies in the cosmic microwave background, its nature still remains unclear [1]. Many models of DM were proposed, in particular, those considering DM to be in the form of Weakly Interacting Massive Particles (WIMPs). Models with inelastic Dark Matter (iDM) and Strongly Interacting Dark Matter (SIDM) were also discussed (see [1] and refs. therein).

In 2020, Pospelov et al. [2] proposed to use nuclear isomers for detection of iDM (and/or SIDM) candidates where possible DM-isomer interaction could lead to induced isomer decay. In experimental searches with ^{180m}Ta [3–5], this process was not observed, and only half-life limits were set (with the best sensitivity $T_{1/2} > 2.9 \times 10^{17}$ yr [5]).

^{178m2}Hf is another possible nuclear isomer to search for iDM (or SIDM) particles. It has high energy of 2.446 MeV, and big half-life of 31 yr related to its high spin 16^+. Normally it decays spontaneously to the ground state of ^{178}Hf by isomeric transition (IT) with emission of cascade of γ quanta with energies up to $\sim$ 600 keV. Possible interactions of ^{178m2}Hf with iDM/SIDM particles 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.

The first experimental search with the ^{178m2}Hf isomer (as an external source relative to a HPGe detector) was carried out in 2023 [6]. Eleven excited levels of ^{178}Hf were considered. No γ peaks, which could indicate the population of those levels, were observed, and the results were presented as limits on the half-life of the ^{178m2}Hf isomer relative to transitions to those excited levels, at the level of $\lim T_{1/2} \sim 10^5$ yr.

In the experiment performed at the INR NASU [7], 17 excited levels were considered; $T_{1/2}$ limits were improved by few times comparing to [6]. They were further improved in the second INR NASU experiment [8], where coincidence technique between a HPGe detector and five NaI(Tl) counters was used.

In all the previous experiments [6–8], measurements were done at the Earth surface. We report here on the first experimental searches for iDM/SIDM induced deexcitations of ^{178m2}Hf isomer performed deep underground (3.8 km of water equivalent) at the Gran Sasso National Laboratory of the INFN. Eighteen excited ^{178m2}Hf levels were considered. These levels were selected due to their high energy (>1 MeV) and significant yield (>25%) of the γ rays emitted; a simplified decay scheme of ^{178m2}Hf is shown in Fig. 1.

2. Experiment and data analysis

In the present experiment, the same ^{178m2}Hf target as in [7, 8] was used. The ^{178m2}Hf was produced by irradiation of a 300 μm thick tantalum foil by 1.2 GeV electron beam during 420 d between 1966 and 1970 at the Kharkiv Institute of Physics and Technology of NASU (see details in [10].) Two ultra-low-background HPGe γ spectrometers were used.

In the measurements with a p-type BEGe detector with volume 118 cm^3, the ^{178m2}Hf source was placed directly on the aluminium endcap. The detector system was shielded with high-purity Cu (5 cm) and Pb (20 cm) and enclosed in a Plexiglas box flushed with high-purity nitrogen gas

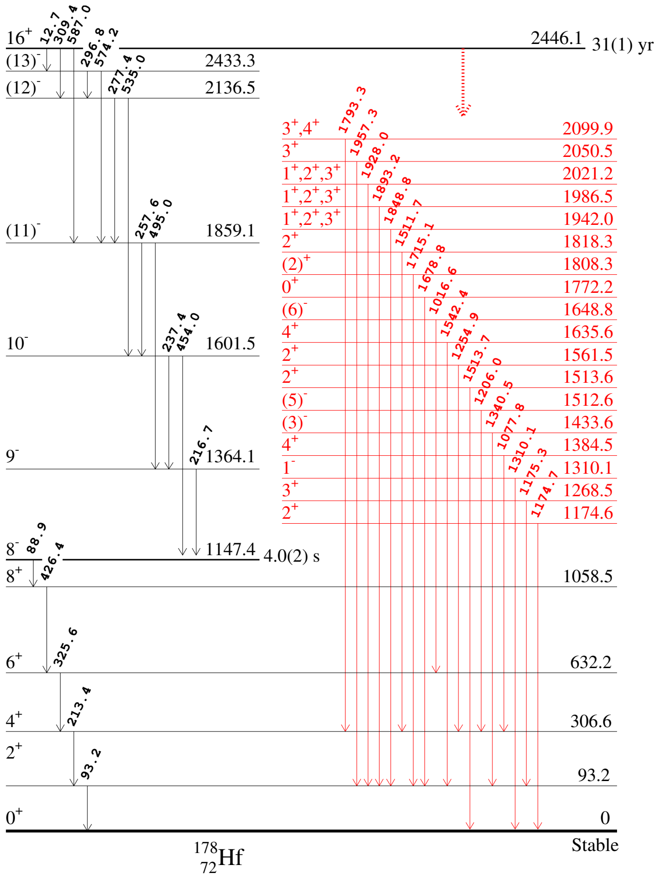

Figure 1: A simplified decay scheme of the ^{178m2}Hf isomer [9]. The allowed spontaneous γ transitions are labelled by black color. Additionally, 18 excited levels of the ^{178}Hf, which might be populated in inelastic scattering of iDM/SIDM particles on the ^{178m2}Hf isomer, constrained by this study, are shown by red.

to reduce as much as possible presence of Rn in the set-up. The energy spectra accumulated with the ^{178m2}Hf source during 558 h and without source during 1660 h are shown in Fig. 2(a). Many summing peaks are present in the ^{178m2}Hf spectrum.

To increase efficiency at higher energies (>1 MeV), the ^{178m2}Hf source was measured with the p-type GeCris HPGe detector with volume 465 cm^3. The source was placed at distance of 22.6 mm from the detector endcap to reduce the summation effects. In addition, Cu plates 2.4 mm thick were installed between the source and the detector to suppress X rays from the source. The detector was shielded by low-radioactive Cu (5 cm) and Pb (25 cm); the system was enclosed in a Plexiglas box flushed with high-purity nitrogen. The energy spectra measured with the GeCris detector during 612 h together with a background spectrum (measured over 1046 h) are shown in Fig. 2(b).

In the spectra with the source, the γ peaks of ^{178m2}Hf prevail below 574 keV; above 574 keV the sum peaks of ^{178m2}Hf and peaks of ^{42}K, ^{44}Sc, ^{60}Co, ^{133}Ba, ^{150}Eu, ^{152}Eu, ^{158}Tb, ^{172}Lu are present. The contribution from the natural radioactivity (^{40}K, U/Th chains) is negligible.

The energy scale and energy resolution of the detectors were calibrated by using clear γ ray peaks present in the spectra.

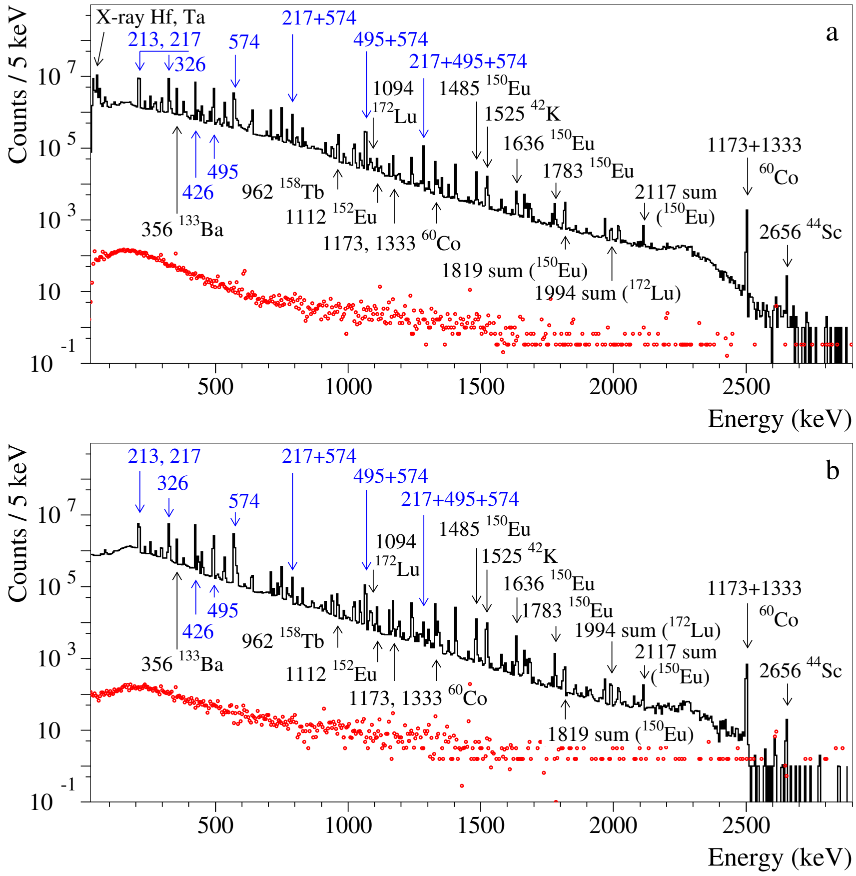

Figure 2: (a) Energy spectra measured with the ^{178m2}Hf source by the BEGe ultra-low-background detector for 558 h and without source over 1660 h (normalized to 558 h), and (b) by the GeCris HPGe detector for 612 h with the source and background data taken over 1046 h (normalized to 612 h). The spectra measured with the ^{178m2}Hf source are drawn by black histograms, while the background data are shown by red circles. The most intensive γ peaks of ^{178m2}Hf are labelled by blue colour. The energy of the γ peaks is in keV.

3. Results and discussion

The total number of peaks above 1 MeV exceeds hundred. Comparing the intensities of the peaks measured with the BEGe (in close geometry) and with the GeCris (far geometry) detectors, the most obvious sum peaks were found. The remaining 61 peaks belong to radioactive impurities with half-lives of a few years induced in the tantalum foil together with the ^{178m2}Hf under 1.2 GeV electron beam. The ^{178m2}Hf activity was derived as 63 Bq while activities of other radioactive nuclides (^{42}K, ^{44}Sc, ^{60}Co, ^{101}Rh, ^{102m}Rh, ^{108m}Ag, ^{133}Ba, ^{150}Eu, ^{152}Eu, ^{154}Eu, ^{158}Tb, ^{172}Lu, ^{174}Lu) were in the range of (0.03 – 23) Bq. The full energy peak (FEP) efficiencies were calculated with the EGSnrc package [11].

We did not find peaks in the data that could be ascribed to the transitions caused by inelastic scattering of iDM or (and) SIDM particles on the ^{178m2}Hf isomer and give only the corresponding half-life limits in approach similar to [6–8]. If an excited level of ^{178}Hf, populated by interaction of iDM/SIDM particles with the ^{178m2}Hf isomer, deexcites via emission of a γ quanta with energy E_γ and a branching ratio b_γ, then:

PoS(MEDEX2025)037

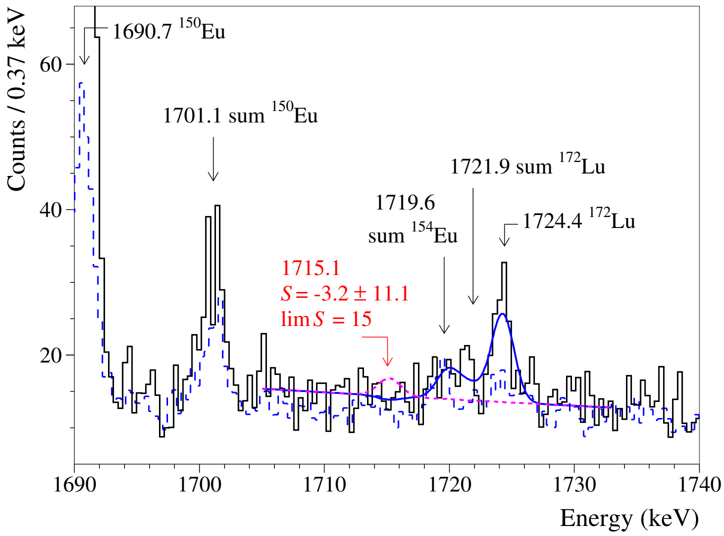

Figure 3: Energy spectrum measured by the GeCris detector for 612 h in the energy region where it is expected a γ peak at 1715.1 keV due to possible interaction of iDM or (and) SIDM candidates with the ^{178m2}Hf isomer (black solid histogram). The fit of the data by an exponential function plus the peak searched for, and three background peaks is shown by blue solid line. The peak with energy 1715.1 keV and area 15 counts, excluded at 90% C.L., is shown by pink dashed line. The energy spectrum measured by the BEGe detector over 558 h, divided by a factor 5 for a better visibility, is drawn by blue dashed histogram. The energy of the peaks is in keV.

$$\lim T_{1/2} = T_{1/2}(^{178m2}\mathrm{Hf})\frac{S_\gamma^{495}\cdot b_\gamma\cdot\zeta_\gamma}{\lim S_\gamma\cdot b_\gamma^{495}\cdot\zeta_\gamma^{495}}, \qquad (1)$$

where: $T_{1/2}(^{178m2}\mathrm{Hf}) = 31$ yr; $\mathrm{S}_\gamma^{495} = 4.0052\times10^6$ is the number of counts in the 495.0 keV γ peak (taking into account the summation with other γ ray transitions in the ^{178m2}Hf decay); $b_\gamma^{495} = 0.715$ is the branching ratio for the transition [9]; ζ_γ^{495} is the FEP detection efficiency for the 495.0 keV γ quanta; $\lim S_\gamma$ is the number of counts in the peak searched for that can be excluded at a given confidence level (C.L.); b_γ and ζ_γ are the branching ratio and the FEP detection efficiency for the γ quanta of interest emitted in the deexcitation of the excited level of ^{178}Hf.

As an example, we show in Fig. 3 the energy spectrum taken by the GeCris detector in the energy region where a peak of 1715.1 keV is expected due to deexcitation of 1808.3 keV excited ^{178}Hf level. The spectrum was fitted in the energy interval (1705 – 1733) keV by an exponential function (to describe the continuous distribution) and by a peak at 1715.1 keV with the proper energy resolution. The model included three background peaks with energies 1719.6 keV (sum peak of ^{154}Eu), 1721.9 keV (sum peak of ^{172}Lu) and 1724.4 (^{172}Lu). The fit (χ^2/n.d.f. = 39/65 = 0.60, where n.d.f. is the number of degrees of freedom) gave the peak's area -3.2 ± 11.1 counts that leads to the $\lim S$ = 15 counts in accordance with [12]. Using eq. (1), this gives a half-life limit $T_{1/2} \geq 5.3\times10^6$ yr.

In a similar way, the limits on the other transitions were obtained, with the $\lim T_{1/2}$ values in the range of $(4 - 64)\times10^5$ yr (preliminary), 1 – 2 orders of magnitude stronger that those in [6, 7] and better (or competitive) with those in [8].

4. Conclusions

The first ultra-low-background search for inelastic scattering interaction of iDM/SIDM particles off the ^{178m2}Hf 31 yr nuclear isomer was performed at the Gran Sasso underground laboratory of the INFN (Italy). Two HPGe detectors, with volume 118 cm^3 and 465 cm^3, were used to measure the ^{178m2}Hf source with activity of near 63 Bq during 558 h and 612 h, respectively. The γ quanta, expected in the deexcitation of excited ^{178}Hf levels that usually are not populated in the spontaneous isomeric decay of ^{178m2}Hf due to the large difference in the levels spin, were not observed. New half-life limits were established in the range of $\sim 4\times10^5 - 6\times10^6$ yr, up to $1-2$ orders of magnitude higher than those obtained in [6, 7] and competitive or better than those in the very recent work [8].

The sensitivity of the experiment was restricted by radioactive impurities present in the ^{178m2}Hf source. Further improvement could be reached after radiochemical separation of hafnium, increase of the ^{178m2}Hf activity and using array of HPGe detectors.

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PoS(MEDEX2025)037