EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Au-198m as novel PAC probe

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Abstract

We propose a test collection and a TDPAC demo experiment with Au-198m ($T_{1/2}$=2.7 d) as potential novel PAC probe. Au-198m is one of the few TDPAC nuclides where the PAC level is populated in an isomeric transition, which is beneficial to minimize after-effects due to chemical change in the decay.

Requested shifts: 3 shifts
Beamline: GLM or GHM + ISOLTRAP MR-ToF MS (CD0)

Among different possible TDPAC (Time Differential Perturbed Angular Correlation) probes [Nagl13], those decaying by Isometric Transitions (IT) are particularly useful in practice, since after-effects due to chemical change of the probe nucleus are minimized. This makes Cd-111m and Hg-199m the TDPAC work horses at ISOLDE and Cu-68m and Pb-204m interesting complements. Au-198m is a potential longer-lived TDPAC nuclide that has not yet been exploited though.

Gold has many technological applications where TDPAC studies could provide important added value. For example Au-Al interfaces are plagued by formation of white plague (Au_5Al_2) or purple plague ($AuAl_2$) that can lead to electrical and/or mechanical failure and TDPAC experiments could help understanding the formation of this plague.

Gold compounds and gold nanoparticles are also promising for various medical applications. For example the FDA-approved drug Auranofin is in clinical use for treatment of rheumatoid arthritis since four decades. Although its' exact mechanism of action remains unknown, it is currently also being studied for cancer therapy or antiviral therapy [Yamashita21]. TDPAC could help to elucidate e.g. binding mechanisms. Au-199, available via the PRISMAP project [PRISMAP], is a promising isotope for therapeutic applications and chelator development for gold could be supported by TDPAC experiments with Au-198m, similar to work with other PAC probes [Kurakina20,Tosato22].

The 12^- isomer Au-198m ($T_{1/2}$=2.3 d) decays with a highly converted M4 transition via the 8^+, 6^+, 5^+, 4^- levels to the 2^- ground state of Au-198 ($T_{1/2}$=2.7 d), see Figure 1. In this sequence the 5^+ level at 312 keV has a half-life of 124(2) ns making it suitable for TDPAC spectroscopy.

Judging from the known quadrupole moments of $11/2^-$ states in neighbouring odd-mass gold isotopes one may expect a quadrupole moment of the order of 1.8 b for the 5^- state. With the known electric field gradient of Au in Cd of $12 \cdot 10^{21}$ V/m^2 [Perscheid83], the expected rotation period would be ≈0.13 µs, i.e. roughly corresponding to one half-life of the intermediate 5^- state, i.e. more than a full period could be observed in a TDPAC experiment.

TDPAC on this state can be performed with the directly populating (204 keV) and depopulating (97 keV) γ-rays. Since the 4^- level ($T_{1/2}$=0.4(2) ns) and most likely also the 6+ level ($T_{1/2}$ unknown, but en passant this experiment will also provide a measurement or at least an upper limit for this number) are short-lived and minimal de-orientation is expected. Hence, the 180 keV γ-ray feeding the 6^+ level can be used as alternative start gate and the 215 keV γ-ray as alternative stop gate leading to 4 possibly exploitable cascades:

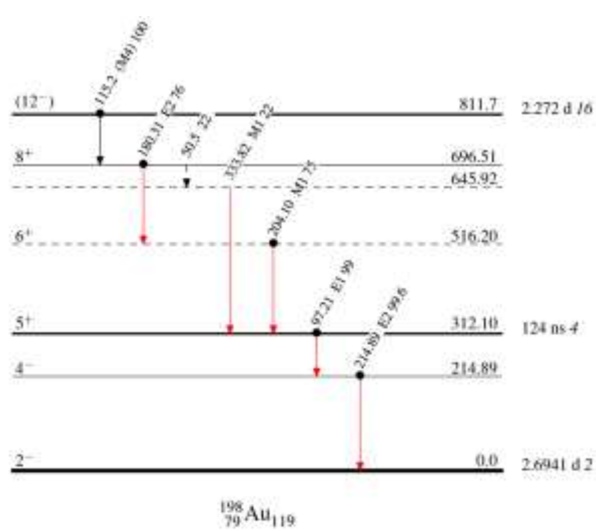

Figure 1: Partial level scheme

The 204 - 97 keV cascade has a combined γγ-intensity of 27% and calculated anisotropies of A_{22} = 0.05 and 0.00 for the other coefficients. The 204 - 215 keV cascade has a combined γγ-intensity of 30% and calculated anisotropies of A_{22} = -0.07, A_{24}=-0.04 and 0.00 for the other coefficients. The 180 - 97 keV cascade has a combined γγ-intensity of 34% and calculated anisotropies of A_{22} = -0.07, A_{42}=-0.011 and 0.00 for the other coefficients. The 180 - 215 keV cascade has a combined γγ-intensity of 38% and calculated anisotropies of A_{22} = 0.10, A_{24}=0.059, A_{42}=0.016 and A_{42}=0.009. The anisotropies of the cascades with 180 keV and 204 keV start γ-rays have opposite sign. To avoid cancellation of the anisotropy due to mixing within the finite energy resolution of the $LaBr_3$:Ce detectors, only the upper part of the 204 keV peak and the lower part of the 180 keV peak will be exploited for gating.

No measured book yields exist for Au-198m at ISOLDE. However, Au-198 beams have been used for a RILIS in-source laser spectroscopy experiment with detection by ISOLTRAP's MR-ToF-MS [Cubiss]. This scan served for calibration purposes of the hyperfine splitting with the well-known 2^- ground state Au-198g. Despite the significantly different hyperfine structure (HFS), also one of the hyperfine peaks of Au-198m could be observed in one tail of the frequency scan. Normalizing to the (near-constant) isobaric Tl-198 intensity, an apparent isomeric ratio Au-198m /Au-198g of about 2:3 could be extracted, i.e. Au-198m represents about 40% of the isotopic Au-198 yield (see Figure 2). Since the absolute transmission to and through the MR-ToF-MS was not measured, the absolute yield of Au-198m has to be estimated differently: FLUKA and ABRABLA simulations predict in-target production yields of $6\text{-}7\cdot10^5$ atoms/μC with standard UC_x targets [Yields]. We assume a release efficiency close to unity for this very long-lived radionuclide. The RILIS ionization efficiency has been determined as >3% [Fedosseev08]. Thus, with 2 μA protons on target an ion beam intensity of $>1.5\cdot10^4$ ions/s Au-198m is expected and within 2.5 shifts a sample of >3 kBq could be collected. To confirm the beam composition and to make sure the collection is performed at the laser frequency assuring maximum yield, we propose to do a RILIS in-source scan on both HFS features of the isomer (a total of about 1 cm^{-1} in range) in combination with the ISOLTRAP MR-ToF MS.

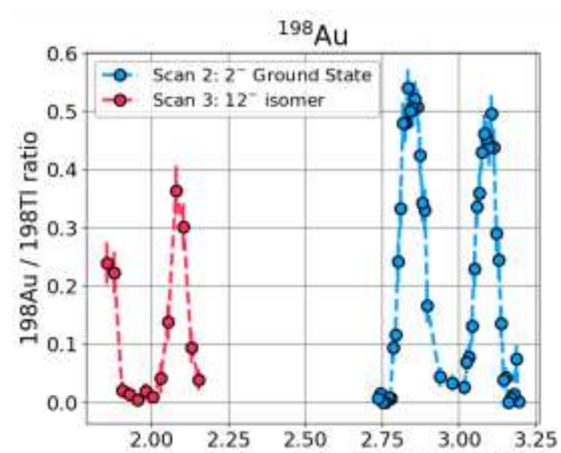

Figure 2: Partial HFS scan of ground and isomeric state

We note that both the HFS scan and the test collection are best performed together with the INTC-P-650 experiment since the same target and ion source unit will be required for both.

Due to the significantly different HFS, only little Au-198g is laser ionized when the RILIS wavelength is optimized for Au-198m. An admixture of Au-198g to the sample activity is not disturbing much since Au-198g decay emits mainly a single 412 keV γ ray, the Compton of which could generate prompt coincidences but not affect delayed coincidences. The same reasoning applies for co-produced isobaric Tl-198 that also emits mainly γ-rays at 412 keV and higher. Moreover, all Tl-198 isomers are short-lived ($T_{1/2}$ = 1.9 h and 5.3 h), i.e. would decay away quickly before the Au-198m TDPAC measurement. Thus, a priori one could expect to obtain a sufficiently clean Au-198m sample.

While such a test sample with few kBq activity will be sufficient for a first demo experiment, the on-line yield of Au-198m at ISOLDE is probably too low for efficient routine use in TDPAC experiments. Higher activities of Au-198m could be obtained by deuteron or alpha induced reactions on platinum targets. After an external irradiation (e.g. at the ARRONAX cyclotron) and radiochemical Au/Pt separation, the sample could serve for off-line mass separation and ion implantation at ISOLDE RILIS. Thus, Au-198m could become in future an interesting "winter physics" beam at ISOLDE.

References:

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[Fedosseev08] V.N. Fedosseev et al., Nucl. Instrum. Meth. B 266 (2008) 4378.

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[PRISMAP] https://www.prismap.eu/radionuclides/portfolio/199Au/

[Tosato22] M. Tosato et al., Appl. Radiat. Isot. 190 (2022) 110508.

[Yamashita21] M. Yamashita, Int. Immunopharmacol. 101 (2021) 108272.

[Yields] https://isoyields2.web.cern.ch/InTargetProductionChart.aspx

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using ISOLTRAP MR-ToF-MS GLM or GHM collection chamber TDPAC setup (building 508)	☒ To be used without any modification ☐ To be modified

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

No additional hazards.