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## The (n, $\gamma$ ) campaigns at EXILL

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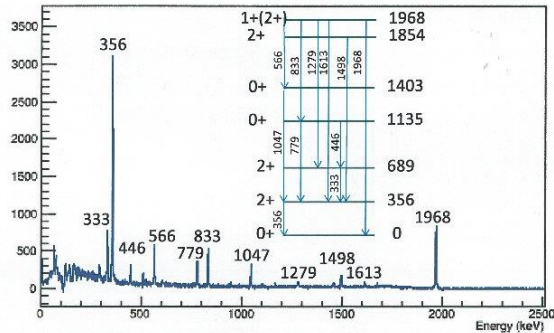
**Abstract.** At the PF1B cold neutron beam line at the Institut Laue Langevin, the EXILL array consisting of EXOGAM, GASP and ILL-Clover detectors was used to perform (n, $\gamma$ ) measurements at very high coincidence rates. About ten different reactions were measured in autumn 2012 using a highly collimated cold neutron beam. In spring 2013, the EXOGAM array was combined with 16 LaBr<sub>3</sub>(Ce) scintillators in the EXILL&FATIMA campaign for the measurement of lifetimes using the generalised centroid difference method. We report on the properties of the set-ups and present first results from both campaigns.

## 1 Introduction

During autumn 2012 and spring 2013 the EXOGAM spectrometer [1] and additional detectors were installed on the high intensity cold neutron guide PF1B [2] of the Institut Laue Langevin (ILL) for the EXILL campaign. EXILL is partially a follow-up of a previous campaign

using 8 EUROBALL capsule Ge detectors and a highly collimated cold neutron beam for (n, $\gamma$ ) measurements [3]. The EXILL campaigns, which took two reactor cycles of 49 days each, used beside the (n, $\gamma$ ) also the (n,fission) reaction on <sup>235</sup>U and <sup>241</sup>Pu targets. An overview of the (n, $\gamma$ ) experiments performed as well as the first results from the (n, $\gamma$ ) reaction on <sup>194</sup>Pt and <sup>195</sup>Pt is presented here.

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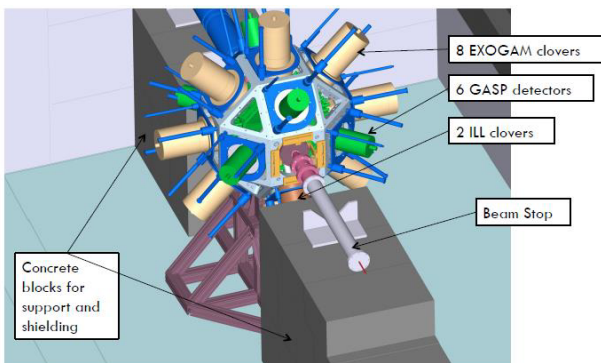


**Figure 1.** Coincident gamma rays in  $^{196}\text{Pt}$  gated by the primary 5953 keV transition showing the deexcitation of the 1968 keV level.

The main advantages of the  $(n,\gamma)$  reaction with thermal or cold neutrons are listed here. Because there is no Coulomb barrier the meV neutrons end up in capture states situated at the neutron separation energies. There is no orbital angular momentum transfer and the capture states have spin  $J+1/2^\pi$  and (or)  $J-1/2^\pi$  with  $J^\pi$  the spin and parity of the ground state of the target nucleus. They generally decay by primary (E1) transitions of several MeV populating, in a statistical way, excited states. By setting gates on these primary gamma rays a given level is selected and all coincident gamma-rays are emitted in the decay of the selected level to the ground state (see Figure 1). Finally, measurements of angular correlations allow the determination of spins and multiplicities.

## 2 The set-ups used

The collimator described in Ref [3] provided, at the target position, a 12-mm diameter cold neutron beam of  $\sim 10^8 \text{ n}/(\text{s}\cdot\text{cm}^2)$ . Perpendicular to the beam, 8 EXOGAM Clover detectors, with their BGO shields, were mounted in a ring while the additional detectors were mounted in the forward and backward directions at  $45^\circ$  (see figure 2). During the first reactor cycle and part of the second one, 6 unshielded GASP Ge detectors and 2 unshielded ILL-Clover Ge detectors were mounted in addition to EXOGAM. A digital data acquisition allowed event rates up to 0.84 MHz to be handled [4].



**Figure 2.** Set-up used for  $\gamma\text{-}\gamma$  and  $\gamma\text{-}\gamma\text{-}\gamma$  coincidences at the PF1B cold neutron beam.

With this set-up the following  $(n,\gamma)$  experiments were done:

- 'High precision measurement of multipole-mixing ratios to test the newly proposed candidate for a low-lying octupole-isovector state in  $^{144}\text{Nd}$ .' M. Scheck (UWS) and T. Kroell (TU-Darmstadt).
- 'A  $\gamma\text{-}\gamma$  angular correlation measurement of  $^{168}\text{Er}$  after  $(n,\gamma)$ .' J. Jolie (Cologne)
- 'Low-spin doorway states feeding the 13/2 high-spin isomer of  $^{195}\text{Pt}$  following  $^{194}\text{Pt}(n,\gamma)$ .' D. Habs (LMU München)
- 'Properties of excited states of nuclei close to  $N=40$  produced in  $(n,\gamma)$  reaction.' A. Korgul (Warsaw)
- 'Search for particle-phonon coupled states in  $^{49}\text{Ca}$  by the reaction  $^{48}\text{Ca}(n,\gamma)^{49}\text{Ca}$ .' S. Leoni (Milano)
- 'Studying three-step gamma cascades following neutron capture in  $^{161}\text{Dy}$ .' M. Krticka (Prague)
- 'Deformation of ground and beta-bands: the  $2^+$  beta to  $0^+$  beta decay in  $^{158}\text{Gd}$ .' Ch. Bernards and V. Werner (Yale)
- 'Measurement of gamma-ray cascades in  $^{78}\text{Se}$ -new information on the Pygmy dipole resonance.' R. Schwengner (Rossendorf)
- 'Identification of the nature of  $3^-$  levels in  $^{96}\text{Mo}$  exploiting the  $^{95}\text{M}(n,\gamma)$  reaction.' M. Scheck (UWS)
- 'Low-spin structure and particle-phonon couplings in  $^{210}\text{Bi}$  studied by using  $^{209}\text{Bi}(n,\gamma)^{210}\text{Bi}$ .' B. Fornal (Warsaw)
- 'The negative parity bands and octupole collectivity: Search for tetrahedral symmetry in  $^{156}\text{Gd}$ .' D. Curien (Strasbourg)

Here we report on results from the  $^{194}\text{Pt}(n,\gamma)^{195}\text{Pt}$  reaction.

During 30 days of the second cycle 16  $\text{LaBr}_3(\text{Ce})$  scintillators from the fast-timing array (FATIMA) collaboration were combined with the 8 EXOGAM detectors for fast-timing measurements. With this set-up the following  $(n,\gamma)$  experiments were done:

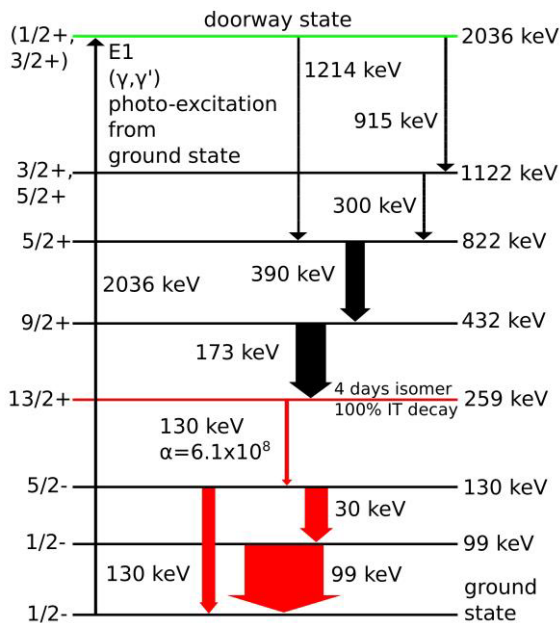
- 'Gamma-ray spectroscopy of  $^{47}\text{Ca}$  by the reaction  $^{46}\text{Ca}(n,\gamma)^{47}\text{Ca}$ .' S. Leoni (Milano)
- 'Low-spin structure and particle-phonon couplings in  $^{210}\text{Bi}$  studied by using  $^{209}\text{Bi}(n,\gamma)^{210}\text{Bi}$ .' B. Fornal (Warsaw)
- 'Test of the  $O(6)$  selection rule in  $^{196}\text{Pt}$ .' J. Jolie (Cologne)

From this campaign we report here on results of the  $^{195}\text{Pt}(n,\gamma)^{196}\text{Pt}$  reaction.

## 3 First results for the $^{194}\text{Pt}(n,\gamma)^{195}\text{Pt}$ reaction

Recently, an alternative production of the platinum isomer  $^{195\text{m}}\text{Pt}$  by resonant  $(\gamma,\gamma')$  photo excitation induced by intense monochromatic gamma beams was proposed

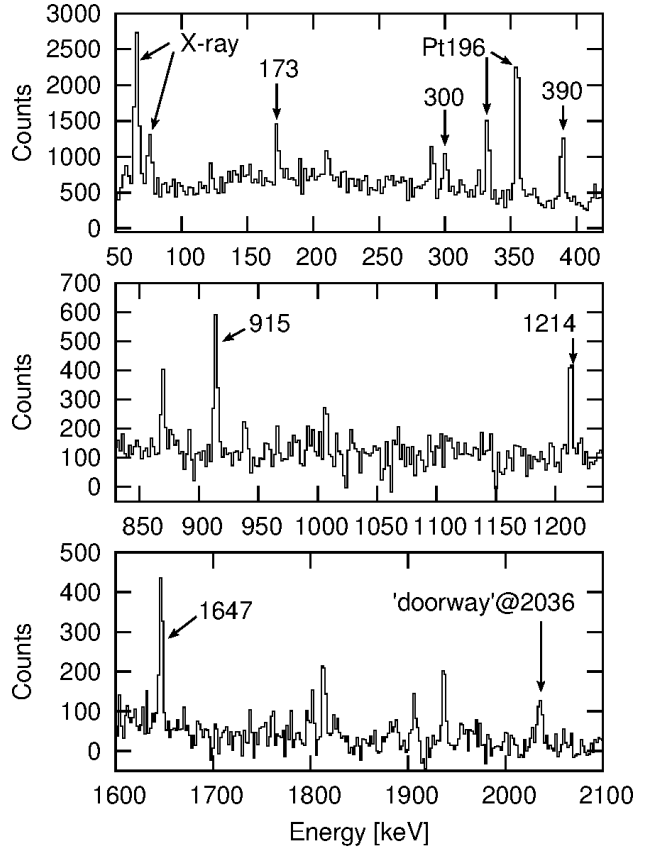
[5]. This method relies on a suitable ‘doorway state’ linking the low-spin ground state with the isomer. The selective population of this isomer is of special interest, as it shows unique properties for diagnostic and therapeutic purposes in nuclear medicine, especially in the treatment of cancer. The  $13/2^+$   $^{195m}\text{Pt}$  isomer has a half-life of 4 days and it intensively emits X-rays (59%  $K_\alpha$  at 66 keV,  $K_\beta$  at 76 keV) and gamma rays (11.7% at 99 keV). Thus, attaching the highly activated  $^{195m}\text{Pt}$  to a suitable biomolecule, it can be used as a tracer for imaging with single-photon emission computed tomography (SPECT) or gamma cameras. Another benefit for the use of  $^{195m}\text{Pt}$  refers to the frequently used platinum based chemotherapeutics like cis-platin or carbo-platin. Here,  $^{195m}\text{Pt}$  could offer a useful tool to assess the tumor uptake during the chemotherapeutical treatment. In addition,  $^{195m}\text{Pt}$  also emits an abundant amount of Auger and conversion electrons, which can be used therapeutically as their energy is deposited in a short range, maximizing the tumor to normal tissue dose ratio. Experimentally, a sample of highly enriched  $^{194}\text{Pt}$  was placed in the very intense cold neutron beam in order to investigate the nuclear structure of  $^{195}\text{Pt}$  via the reaction  $^{194}\text{Pt}(n,\gamma)^{195}\text{Pt}$  and search for such ‘doorway states’.



**Figure 3.** Partial level scheme of  $^{195}\text{Pt}$  showing the relevant isomeric transitions (red), the isomer populating transitions (black) and the idea of  $(\gamma,\gamma')$  induced activation of the isomer  $^{195m}\text{Pt}$  from the ground state of  $^{195}\text{Pt}$  via the doorway state (green) (color online).

In a still ongoing analysis the level scheme is being investigated using coincidence measurements and angular correlations with special regard to the isomer populating pathways. Therefore, high energy primary transitions decaying from the neutron capture state at 6.105 MeV, especially to the strong isomer populating starting at the 1122-keV level with a cascade of 300 keV, 390 keV, 173 keV transitions, which first has been observed by D. D. Warner *et al.* in 1982 [6], were investigated. A ‘doorway

state’ in  $^{195}\text{Pt}$  with an energy of 2.036 MeV was found, which is populating the isomer  $^{195m}\text{Pt}$  and, in addition, directly decays into the ground state fulfilling the requirements of gamma ray induced activation of  $^{195m}\text{Pt}$  (see figure 3, 4).



**Figure 4.** Background corrected coincidence spectrum of the relevant energy ranges in  $^{195}\text{Pt}$  gated on the primary transition of 4.069 MeV leading to the ‘doorway state’ at 2.036 MeV. The upper figure shows the strong isomer populating transitions of the ‘Warner’s cascade’; the middle figure shows two transitions depopulating the ‘doorway’ and feeding the cascade; the lower figure shows the doorway state decaying directly into the ground state (cf. fig. 3).

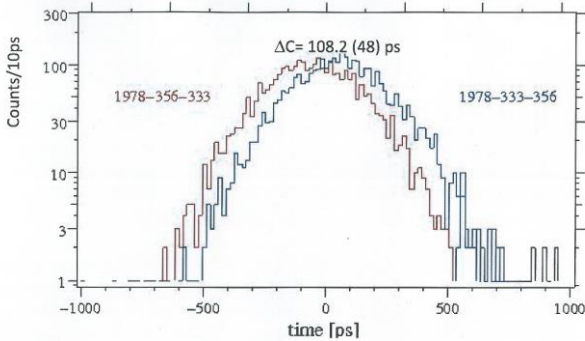
Moreover other promising  $\gamma$ -transitional pathways leading to the isomer were observed.

## 4 First results for the $^{195}\text{Pt}(n,\gamma)^{196}\text{Pt}$ reaction

Many years ago, the even-even nucleus  $^{196}\text{Pt}$  was proposed as an example of the SO(6)-limit of the Interacting Boson Model (IBM) [7]. When the Hamiltonian for  $N$  s,d bosons is composed by the linear combination of second order Casimir operators of the group chain:  $\text{SO}(3) \subset \text{SO}(5) \subset \text{SO}(6) \subset \text{U}(6)$ , the quantum numbers classifying the irreducible representation (irrep) of these groups:  $L, \nu, \sigma, N$  are valid and the Hamiltonian is analytically solvable. The lowest states have  $\sigma=N$ . States at higher energies can have



$\sigma = N-2, N-4, \dots$ . Recently, the need to test the SO(6) symmetry and not only the SO(5) properties in  $^{196}\text{Pt}$  was stressed [8]. When testing the goodness of the description of an SO(6) nucleus one thus should look for properties involving states with different  $\sigma$ . One of these properties concerns the electric quadrupole transition operator. In the SO(6) limit, the E2 transition operator is a generator of SO(6). Now, generators cannot act outside their irrep and the strict selection rule  $\Delta\sigma = 0$  follows. In order to test this selection rule, absolute B(E2) values between  $\sigma = N-2$  and  $\sigma = N$  states need to be measured. This is not an easy task as the first  $\sigma = N-2$  states in  $^{196}\text{Pt}$  are at high energy and have low spins. They are not populated by standard in-beam reactions with ions, but they are by neutron capture. It is, therefore, not an accident that Ref [7] relied on ILL data. In 1990 an attempt was made to measure the lifetime of the lowest  $\sigma = N-2$  state at 1402.7 keV using the GRID method [9] at GAMS4 [10]. For this  $0_3^+$  state a lower lifetime limit of  $\tau > 1.8$  ps could be established showing that the B(E2) towards the  $2_1^+$  was smaller than  $0.034 e^2b^2$  and, as such, at least an order of magnitude smaller than allowed transitions between states with  $\sigma = N$ .

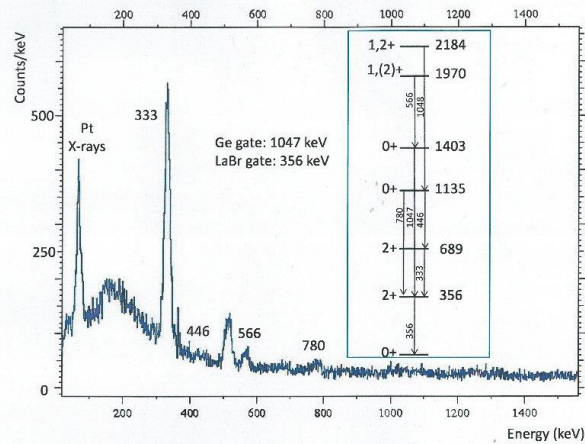


**Figure 5.** The binned time spectra using the 356 keV ground state transition as start for the TAC (left curve) and once as stop (right curve) (color online).

Using the FATIMA and EXILL set-ups triple coincidences can be used in which a Ge gate selects the cascade of gamma-rays to be measured by the LaBr<sub>3</sub>(Ce) scintillators. These fast signals can then be used for the generalized mirror symmetric centroid difference method [11,12] to measure lifetimes. The prompt response difference curve (PRD) was obtained using an  $^{152}\text{Eu}$  source and transitions in  $^{49}\text{Ti}$  after neutron capture [13,14]. The method can be used to measure lifetimes down to about 10 ps.

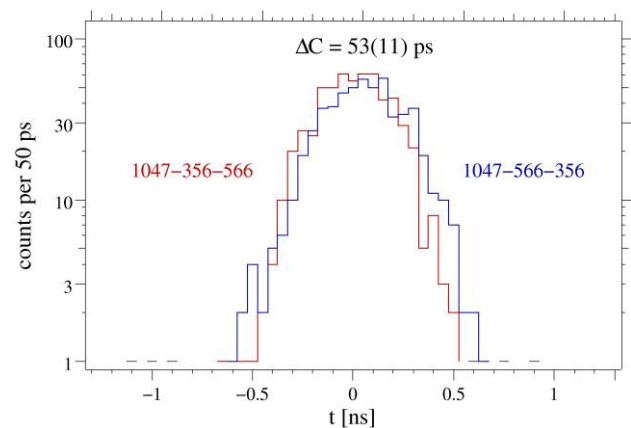
The experiment took place during 19 hours. Because the thermal cross section for  $^{195}\text{Pt}(n,\gamma)^{196}\text{Pt}$  is 28 barns and dominates over those of the other Pt isotopes, a  $^{nat}\text{Pt}$  target of 143 mg was used. The Ge total count rate was 263 kHz and the LaBr<sub>3</sub> one 192 kHz. In total  $3 \times 10^{10}$  events were measured. Figure 5 shows the time spectra of the 333-356 keV cascade gated by the 1978-keV transition in  $^{196}\text{Pt}$ . The observed centroid difference between both spectra corresponds to  $\Delta C = 108(5)$  ps. Using the PRD curve and  $\Delta C = \text{PRD}(333) - \text{PRD}(356) + 2\tau$  together with  $\text{PRD}(333) - \text{PRD}(356) = 8(10)$  ps [12] a

lifetime of  $\tau(2_1^+) = 50(6)$  ps is deduced which agrees very well with the literature value of 49.2(2) ps. Being confident that the method works, the lifetime of the 1402-keV  $0_3^+$  state was then measured. Again the 356-keV transition provided the decay signal for the generalized centroid difference method.



**Figure 6.** Relevant part of the level scheme of  $^{196}\text{Pt}$  (insert) and the double-gated LaBr<sub>3</sub>(Ce) spectrum.

The feeding signal was obtained from the 566-keV transition populating the 1402-keV state. The Ge gate was set on the 1048-keV transition of the decay towards the first excited state. Figure 6 shows the double-gated LaBr<sub>3</sub>(Ce) spectrum. The result is shown in Figure 7. The effective lifetime deduced from the centroid shift  $\Delta C = 53(11)$  ps using  $\text{PRD}(566) - \text{PRD}(356) = -54(10)$  then yields  $\tau_{\text{eff}} = \tau(2_1^+) + \tau(0_3^+) = 54(7)$  ps leading to an upper limit for the lifetime of  $\tau(0_3^+) < 12$  ps. Using also the lower limit from [10] we find that  $0.75 \text{ W.u.} < B(E2; 0_3^+ \rightarrow 2_1^+) < 5 \text{ W.u.}$  and  $0.06 \text{ W.u.} < B(E2; 0_3^+ \rightarrow 2_2^+) < 0.41 \text{ W.u.}$  Clearly no collective B(E2) values are found between the states, confirming the validity of the SO(6) symmetry. A recent Gammasphere experiment using multiple Coulomb excitation confirmed our result with  $0.62 \text{ W.u.} < B(E2; 0_3^+ \rightarrow 2_1^+) < 4.9 \text{ W.u.}$  [15].



**Figure 7.** The binned time spectra using the 356 keV ground state transition as start for the TAC (left curve) and as stop (right curve) (color online).

## 5 Conclusion

The EXILL campaigns have provided a huge amount of new data using a high intensity cold neutron beam and the  $(n,\gamma)$  and  $(n,fission)$  reactions. Here we reported on the  $(n,\gamma)$  experiments from the EXILL and EXILL&FATIMA campaigns and showed promising first results on  $^{195}\text{Pt}$  and  $^{196}\text{Pt}$ . The success of the campaigns has led to the proposal of a dedicated new ILL instrument for prompt fission studies using gamma-ray spectroscopy called FIPPS [16]. The option of using also the  $(n,\gamma)$  reaction at FIPPS is important.

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