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High resolution neutron (n,xn) cross-section measurements for ^{206,207,208}Pb and ^{209}Bi from threshold up to 20 MeV

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Abstract. Gamma production cross sections for neutron inelastic scattering reactions (n,xn) (x = 1, 2, 3) were measured for three different highly enriched targets of ^{206}Pb, ^{207}Pb and ^{208}Pb and for ^{209}Bi. Using the known decay schemes of these isotopes, the total inelastic cross sections and level inelastic cross sections were determined. The measurements are continuous in neutron energy and cover a wide interval (from threshold energy up to about 20 MeV). The experiments were performed at the GELINA white neutron source, at the 200 m flight path station with a time resolution of 8 ns, resulting in an unprecedented neutron energy resolution of 1.1 keV at 1 MeV (36 keV at 10 MeV).

1 Introduction

Lead and bismuth are interesting materials from the point of view of a new generation of nuclear reactors and also for designing innovative Accelerator Driven Systems (ADS) in which they can be used both as target and coolant. The impact of nuclear data uncertainties on transmutation of actinides in ADS was assessed in [1]. However, the existing data concerning neutron inelastic scattering are often not in good agreement with each other, not to speak about the lack of (especially continuous) data in some energy regions. Taking into account the drastic energy change after an inelastic interaction and the fact that inelastic cross sections amount in some energy regions to about 30% of the total cross section we decided to make these measurements with a good accuracy and a good energy resolution in a wide energy range. The here reported measurements were obtained by recording the emitted gamma rays following the inelastic reaction in time of flight (TOF) mode. The technique is described in detail in [2] which indicates also the detector's disposal for convenient and precise angle integration.

2 Experimental layouts

The measurements for Bi were done with two high purity (HP) Ge detectors. Subsequently, for the measurements of lead isotopes, the system was upgraded to three Ge detectors (for ^{207}Pb), then to four detectors and a digitised acquisition system doubled the classical, analogue one. The neutron beam was collimated to a diameter of 6 cm and a lead wall shielded the detectors from the collimation system. The beam monitoring was achieved by an ionization chamber with ^{235}U deposits (see [2] for details). For each measurement the detection efficiency of Ge detectors was determined with a calibrated ^{152}Eu point source and then the detection geometry was fine tuned in a MCNP simulation in order to precisely reproduce the measured data with the calibration source. Subsequently, using the same geometry, a new MCNP calculation was done for the extended geometry of the target intercepted by the neutron beam. The total measuring time for one isotope varied from two to three months. Data acquisition (DAQ) was done using the standard GELINA acquisition system with ADCs for amplitude and a multi hit time coder operated in coincidence for the time information. For the measurements of lead isotopes, a new DAQ based on fast digitizers was used in parallel with the classical one. The analysis of the two sets of data yielded results in remarkable agreement with each other (see ref. [3] for details on the upgraded system). The time resolution of Ge detectors was carefully tuned to achieve 5 ns FWHM. The DAQ system recorded for each event the amplitude coded with 12 bits and time with a width of 8 ns/channel.

3 Data presentation

In the following, the essentials of the results for each isotope will be presented, together with some particularities concerning each individual decay scheme and a short discussion and comparison with calculations performed with the default version of the TALYS code and with other measurements, when existing. The measured quantities, and this is valid for all measured isotopes, were the gamma production cross sections for various levels as a function of incident neutron energy. Following the procedure described in [2], from these quantities, using the known decay scheme, total inelastic cross sections and individual level cross sections were obtained.

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3.1 ^{209}Bi inelastic and (n,2n) reactions

Two Ge detectors placed at 110° and 150° with respect to the beam were used in the experiment. Preliminary data for Bi have already been presented at the previous ND conference [4]. Here the accent will be on some specific aspects and on the results obtained for the (n,2n) channel. Considering the decay scheme of ^{209}Bi, for each level up to 3.46 MeV, at least one transition was observed and measured. A particularity of the decay spectrum of ^{209}Bi is that besides the 896.28 keV level, there is another level at 895.9 keV coming from the (n,2n) channel and that opens above 9 MeV. The two levels can not be separated and therefore both the inelastic and (n,2n) cross sections will be affected. Cross section estimates obtained with the TALYS code indicate a maximum of 35 mb for the (n,2n) reaction at 12 MeV which is about 50% of the measured gamma production cross section of the inelastic level (896.28 keV) at that energy. Gamma production cross sections were measured for a total of 39 transitions. Their contribution to the total inelastic cross section diminishes with the increasing excitation energy of the level. TALYS calculations give a reasonable agreement with gamma production cross section of most intense lines; for higher lying levels, the agreement is variable and anyway the TALYS calculation for individual transition only extends up to 3.13 MeV. The total inelastic cross section build from gamma production cross sections is shown in figure 1 together with a TALYS calculation (with default parameters, version 0.57) and some other existing data. A slight disagreement with TALYS calculation can be observed: TALYS underestimates data both for low and high energies. Moreover, the dip in the cross section around 7 MeV is not reproduced. In the same energy region other data are above ours. Looking up in the decay scheme one can observe for these energies many levels decaying directly to the ground state. In an effort to clarify this situation, a new measurement was done with a reduced gain, such as to rend observable possible high energy gamma lines. Even if the strength would be distributed in energy and therefore not enough to see individual lines, one should be able to observe at least a wide structure for an appropriate selection in incident neutron energy, especially that the background in this energy region is almost negligible. The difference between our measured inelastic cross section and the TALYS calculation at $E_n = 7$ MeV amounts to about 250 mb. If the difference would come from the contribution of direct decays to the ground state, then these decays should be observed in the experiment despite low detection efficiency. Another test was done by using BaF_3 detectors of large volume instead of Ge detectors, to enhance detection efficiency: again the result was negative. All these tests gave us confidence in the measured cross sections.

The (n,2n) reaction has a threshold at 7.495 MeV; therefore, only data corresponding to higher incident neutron energies were scanned for building the gamma production cross sections for transitions belonging to this channel. The first two levels, at 65.5 keV and 510.5 keV could not be observed: the first one is strongly converted and is anyway below our registration threshold, while the second can not be distinguished from the annihilation line. A total of eight transitions were measured. They do not include the 895.9 keV transition

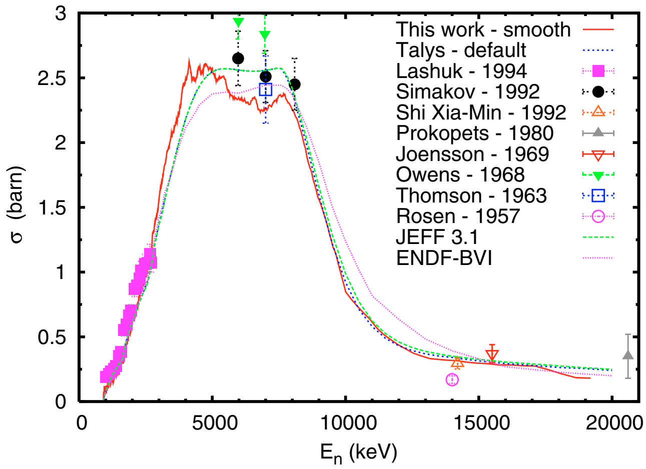

Fig. 1. Total inelastic cross section for ^{209}Bi.

from the level at 959.3 keV in ^{208}Bi as this one overlaps a (n,n′) transition, as mentioned previously. The values of the gamma production cross sections for these transitions at their maximum (which is normally about 2 MeV above threshold) are comprised between 60 mb and 210 mb. TALYS calculations were available for the first 5 of the 8 measured transitions; their agreement with the measurements is rather poor. No other data were found in the literature for the (n,2n) reaction.

3.2 ^{206}Pb inelastic and (n,2n) reactions

The measurements were accomplished by using four Ge detectors placed at 110° and 150° to the left and to the right of the incident beam using a target of 99.82% purity. The (n,n′γ) technique used in the present measurements encounters for ^{206}Pb two difficulties. Firstly, one transition, from the second excited state to the ground state is an E0, therefore not observable with our detectors. Secondly, the level at 2200.14 keV is an isomeric level with a life time of 125 μs. This is 5 times more than our measured time interval. Therefore the decay lines of this isomer, 516.18 keV and 202.44 keV could not be observed in the prompt spectrum. On the other hand, the lifetime is 10 times smaller than the distance between two consecutive neutron bursts. As a consequence, the isomer almost completely disappears between bursts and the observation of the decays fed by the isomer could be done with good resolution for the incident neutron energy. The absence of any detectable yield below the inelastic threshold of the 880.98 keV transition, that is fed by the 516.18 kev decay line of the isomer is a proof that the isomer decays practically completely between bursts. Figure 2 shows the gamma production cross section for the 803.06 keV transition contributing essentially to the total inelastic cross section. The figure also shows other data and three TALYS calculations: the first is one with default parameters; the measured data are clearly overestimated. The second is a calculation from which the isomer contribution was deduced and the third is a calculation with both contributions deduced: of the isomer and of the E0 transition. Already the second calculation is very close to

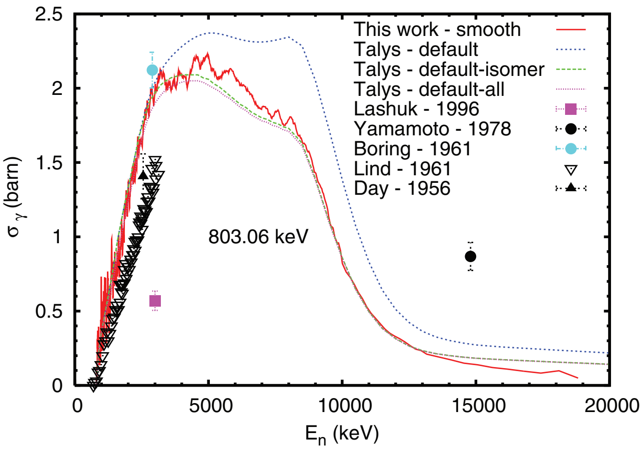

Fig. 2. Gamma production cross section for the 803.06 keV transition in ^{206}Pb.

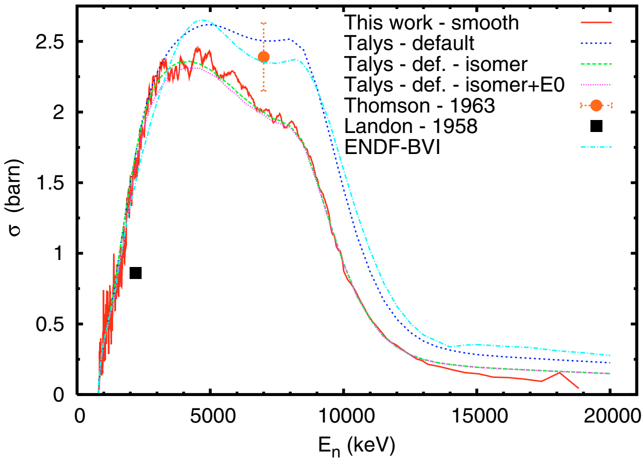

Fig. 3. Total inelastic cross section for ^{206}Pb.

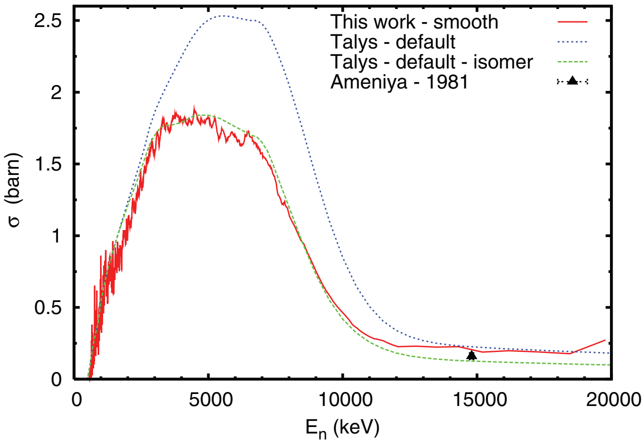

Fig. 4. Total inelastic cross section for ^{207}Pb.

measurement. Visibly, the E0 contribution is almost negligible which would have to be expected for a level with a structure neatly differing from that of its neighbours. A close up view at the low energies shows prominent resonant structures, the observation of which is possible due to the very good energy resolution of the measurements. Other 22 transitions from the decay of excited levels up to 3.56 MeV were measured. Up to 2.2 MeV, for each excited level at least one transition was observed. Based on the known level scheme of ^{206}Pb, the total inelastic cross section was built, as well as level cross sections (for levels below 2.2 MeV). The total inelastic cross section is shown in figure 3 together with TALYS calculations similar to that in figure 2 and with some data from the literature. The total inelastic cross section lacks the contribution from the isomeric level and from the E0 transition. Apart from these contributions, it is precise up to 3.2 MeV, above this limit the proposed values are only lower limit estimates. However, the observed strong decrease of the strength carried by the higher lying levels with the increasing neutron energy indicates that the measurement should be fairly close to reality. The gamma production cross section of two rays from the (n,2n) channel, namely 573.88 keV and 703.44 keV were measured. They amount to 100 mb and 300 mb respectively, at their maximum (above 12 MeV). TALYS calculations underestimate and respectively overestimate these measurements. After upgrading the experimental setup [2], a new set of measurements was started, using a new target of 88.5% purity (^{207}Pb being the main impurity), 7 cm diameter (i.e., larger than the beam diameter of 6 cm) and 5 mm thickness. Also the distances of the detectors changed and the DAQ was entirely based on fast digitizers. The non negligible amount of ^{207}Pb impurity required the application of adequate corrections obtained from a previous measurement of this isotope (see below). The agreement with the previous results was within 1–2%, i.e., better than the assigned uncertainty of the previous set of data (5%). The better statistics of this last measurement – mostly due to the much thicker target – allowed to observe and measure 4 new transitions for the (n,n′γ) channel and one new transition (987.66 keV) for the (n,2n) channel. The agreement of the two sets of data gave us full confidence in the new measuring setup.

3.3 ^{207}Pb inelastic and (n,2n) reactions

This experiment, anterior to that on ^{206}Pb, was accomplished with three Ge detectors. The used target contained 2.16% ^{206}Pb and 5.48% ^{208}Pb, i.e. non negligible amounts. As a consequence, corrections were necessary to account for their contribution in the region where (n,2n) and (n,3n) channels were open. Besides, like in the case of ^{206}Pb, an isomeric level (1633.36 keV) with a lifetime of 0.8 s made the analysis more complex. The lifetime of the isomer is much larger than the distance between two consecutive bursts (1.25 ms); therefore, its prompt decay was negligible during the measuring time (first 20 μs). The present measurements do not include the isomer contribution and its contribution was extracted also from the TALYS calculations in order to compare them to the experiment. The gamma production cross section was measured for 15 γ-rays spanning excitation energies up to 4317 keV. Figure 4 shows the total inelastic cross section of ^{207}Pb together with TALYS calculations (default parameters) and other data, when existing. The main contribution to this cross section comes from the first two transitions, 569.7 keV and 897.8 keV. The agreement with TALYS (after deducing the isomer contribution) is fairly good. For the low energy region (below 2 MeV), prominent resonant structures are

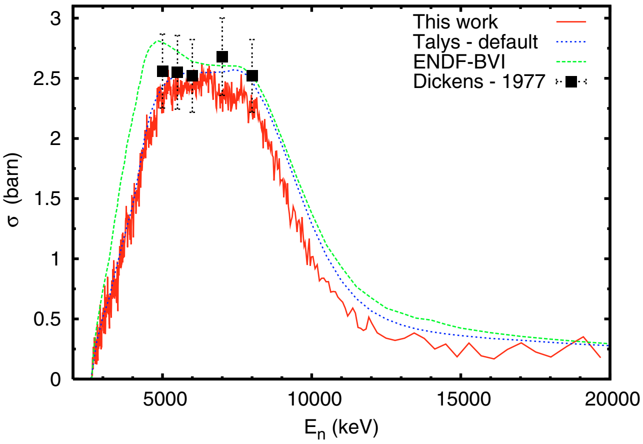

Fig. 5. Total inelastic cross section for ^{208}Pb.

observed, as expected for a doubly magic compound nucleus. For the (n,2n) channel, four transitions were measured for the first time: 808.3 keV, 537.47 keV, 880.98 keV and 1704.45 keV. The first one is the most important, with a cross section of about 1.1b from 10 MeV to 15 MeV.

3.4 ^{208}Pb inelastic, (n,2n) and (n,3n) reactions

The sample used had an isotopic purity of 88.1%. The main contaminants were ^{206}Pb (11%) and ^{207}Pb (0.9%) and therefore corrections (obtained from the previous measurements) were applied to the measured gamma production cross sections. The simple decay scheme of this doubly magic nucleus and the absence of isomers, low energy transitions or E0 levels allowed using only three transitions to build the total inelastic cross section: 2614.5 keV, 4085.4 keV and 4229.5 keV. The obtained results are shown in figure 5 together with TALYS calculations and other existing data.

Five transitions from the (n,2n) channel were measured of which 595.7 keV one has the largest cross section. In comparing with TALYS calculations, again the contribution from the isomeric state in ^{207}Pb had to be subtracted, as the mentioned level is fed by the isomer. After this operation, the agreement of the measurement with calculation is good. The same good agreement is observed for the 897.78 keV level, this time corrections for the isomer not being necessary (level not fed by the isomer). Another interesting situation occurred for the 1593 keV transition from the same channel: though this channel opens above 10 MeV, it was observed at much lower energies. In reality, the observed line was the double escape of the main, most important transition of 2614.5 keV. At the opening of the channel for the 1593 keV transition and at higher energies, the contribution from the double escape of the main transition became already insignificant. For the (n,3n) channel, the main transition was measured, 803.06 keV. The data are in fair agreement with a previous measurement of Vonach et al. [5] and with TALYS calculations.

4 Conclusions and perspectives

A large amount of data was obtained for Bi and the main three isotopes of lead. These data are: gamma production cross sections (basic result), total inelastic cross sections and level cross sections (derived quantities, by using known level schemes) for the inelastic and (n,xn) channels. These data are continuous in energy with a very good resolution and covering a wide range (up to 20 MeV). The uncertainty is generally below 5% in the low energy range (below 10 MeV) and when statistics were not enough, few time channels were taken together to ensure reasonable uncertainties. The reported data are new. The reliability was checked in the particular case of ^{206}Pb by re-measuring with another setup. Data were compared with TALYS calculations (always with default parameters in order to check their predictive value) and with other data, when available. The employed method, based on knowledge of the level scheme may show for some (generally few) particular situations certain weaknesses. The experimental setup was continuously upgraded, becoming a veritable "data factory".

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