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ガンマ線分光法による核データ測定精度の高度化に関する研究

（研 究 報 告）

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（研究報告）

古高和禎

要旨

本報告は、著者が核燃料サイクル開発機構において、平成9年11月から平成12年10月までの期間に博士研究員として行った研究内容をまとめたものである。

本報告は、二つの内容に分かれる。すなわち、一つは、熱中性子吸収断面積の測定の高度化に関する研究である。今一つは、HHS検出器を用いた光核反応断面積の微細構造測定の高度化に関する研究である。

1) 放射化法を用いたγ線測定による熱中性子吸収断面積測定において、得られる結果の精度に影響を及ぼす主な要因には、γ線収量の統計精度の他に(1)γ線ピーク検出効率の校正精度、及び(2)γ線放出率の精度があげられる。本研究では、高速三次元同時計測システムを作成することにより、(1)γ線ピーク検出効率を精密に校正するための、γ-γ同時計測法を用いた標準γ線源放射能の精密測定、及び(2)短寿命核のγ線放出率の精密測定に用いるための、β線検出器にプラスチックシンチレータを用いたβ-γ同時計測法の開発及び、それを使用した ^{100}Tc のγ線放出率の精密測定を行い、熱中性子吸収断面積測定の高度化を図った。
2) 熱中性子吸収断面積が小さい核種に対しては、巨大共鳴領域のγ線を用いた光吸収反応による核変換が提案されている。光吸収反応による核変換を効率的に行うためには、光吸収断面積の入射γ線エネルギー依存性を詳細に知る必要がある。本研究では、高分解能高エネルギーγ線スペクトロメータ(HHS)を用いた光吸収断面積の微細構造測定をより精密で信頼できるものとするために、精密なモンテカルロシミュレーション計算を実施し、検出器の標準γ線応答関数の整備を行った。

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A study on improvements in accuracy of nuclear data measurements using γ–ray spectroscopic methods

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abstract

This report describes the study done by the author as a postdoctoral research associate at Japan Nuclear Cycle Development Institute.

This report is divided into two parts: improvements in accuracy in determination of thermal neutron capture cross sections, and improvements in accuracy of photo-nuclear absorption cross section measurements using the HHS.

1) In the measurements of thermal neutron capture cross sections using an activation method, accuracies of the final results attained are limited by (1) accuracy of γ–ray peak detection efficiencies, and (2) accuracies of γ–ray emission probabilities. In this study, to determine thermal neutron capture cross sections more accurately, the following researches have been done using a newly developed three-dimensional coincidence measurement system: (1) accurate determination of γ–ray standard sources using a γ–γ coincidence method, for precise calibration of γ–ray peak detection efficiency, and (2) development of a β–γ coincidence measurement system using a plastic scintillation detector as a β–ray detector, for the determination of γ–ray emission probabilities of short-lived nuclides, and measurement of γ–ray emission probabilities of ^{100}Tc nuclide using the coincidence system.

2) To transform radioactive nuclides with small thermal neutron capture cross sections, use of photonuclear absorption reaction has been suggested. In order to transform these nuclides efficiently using the reaction, one has to know detailed behavior of the photo-absorption cross sections. In this study, a Monte-Carlo simulation code has been used to create a standard set of γ–ray response functions of the high-resolution high-energy spectrometer (HHS), to enable reliable analyses of the data obtained by the spectrometer.

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第１章 研究の概要

1.1 熱中性子吸収断面積測定の高度化

放射性核種の核反応断面積などの核データを高精度で測定することは、高レベル放射性廃棄物の分離核変換方法を確立する上で非常に重要である。特に、中性子を用いた合理的な分離核変換方法を検討するためには、高精度の熱中性子吸収断面積が必要となる。

従来より、熱中性子吸収断面積の測定は、放射化法を用いて行われてきている。この方法では、断面積を測定したい試料を中性子で照射し、放射化された試料から放出されるγ線を測定する。そして、γ線のエネルギーに対応したピーク収量から放射化された試料の量を算出し、断面積を求める。この際、得られる断面積の精度は主に次の要因によって制限される:

(1) γ線検出器がピークとして検出される確率(γ線ピーク検出効率)

(2) 放射性核種が崩壊する際にγ線を放出する確率(γ線放出率)

前者、すなわちγ線ピーク検出効率は、放射能が精度良く決められた標準γ線源を用いて校正することにより精度良く決定することが出来る。しかしながら、市販の標準γ線源の放射能は大抵数％、良いものでも2％程度の精度でしか決定されていない。したがって、これらの線源を用いてγ線ピーク検出効率を高精度で決定するためには、線源の放射能を高精度で決定しなおす必要がある。

一方、γ線放出率は、従来4π β−γ同時計測法を用いた高精度測定が行われてきた。半減期が数分程度よりも長い核種に対しては、同方法を用いることにより、γ線放出率を1％以下の精度で決定することが出来る。しかしながら、それよりも短寿命の核種に対しては適用が困難であり、他の手法を用いることが必要となる。

本研究では、高速の三次元同時計測システムを開発することにより、(1)γ−γ同時計測法を用いて標準γ線源の放射能を高精度で決定してγ線ピーク検出効率の高精度校正に用いること、及び(2)β線検出器にプラスチックシンチレータを用いたβ−γ同時計測法の開発を行い、その有効性検証を実施すると共に、核変換対象核種として重要な^{99}Tcの(n,γ)反応生成核種である^{100}Tcのγ線放出率精密測定に応用した。

(1) 同時計測法による^{60}Co標準γ線源の放射能の高精度測定

開発した同時計測システムの性能を確認するために、γ−γ同時計測法により市

販の ^{60}Co 標準γ線源の放射能を高精度で測定する実験を行った。この実験から、同線源の放射能が 135.4±5.8 kBq と求まり、経時減衰の補正をしたカタログ値と誤差の範囲で一致したので、同システムの有効性が確認された。詳細は別添えの報告書(第 2 章、2.1)としてまとめた(1998 年核データ研究会において発表)。以後、γ-γ同時計測法による測定を継続し、同 ^{60}Co 標準γ線源の放射能を 0.48%の精度で決定した。

(2) プラスチックシンチレータを用いた同時計測法の短寿命核のγ線放出率の精密測定への有効性の検証

β線検出器にプラスチックシンチレータを用いたβ-γ同時計測法の、短寿命核のγ線放出率の精密測定への有効性を検証するために、放出率が既知(100%)の ^{28}Al(半減期 2.2 分)の 1779 keV γ線の放出率測定を行った。解析の結果、^{28}Al の 1779 keV γ線の放出率が 0.995±0.014 と 2%以下の精度で求まり、短寿命核のγ線放出率の精密測定に対して同手法が有効であることが検証できた。詳細は別添えの報告書(第 2 章、2.2 及び 2.3)としてまとめた。(前者は 1999 年核データ研究会において発表。後者は *Journal of Nuclear Science and Technology* 誌に掲載)

(3) ^{100}Tc のγ線放出率の精密測定

^{99}Tc は、半減期が長く(21 万年)収率が比較的大きな(約 6%)核分裂生成核種であるが、地中での易動性が指摘されており、中性子を用いた核変換の候補にあげられている。中性子を用いた核変換を検討するには、熱中性子吸収断面積を精密に知る必要がある。しかし、^{99}Tc の中性子吸収反応の生成物である ^{100}Tc は、半減期が 15 秒と非常に短いためγ線放出率が精密に決定されておらず(誤差 17%)、^{99}Tc の熱中性子吸収断面積は精密に決定されていない。そこで、β線検出器にプラスチックシンチレータを用いたβ-γ同時計測法を用いて、^{100}Tc のγ線放出率の精密測定実験を行った。詳細は別添えの報告書(第 2 章、2.4)としてまとめた。(日本原子力学会 2000 年秋の大会にて発表)

1.2 光核反応断面積の微細構造測定の高度化

熱中性子吸収断面積が小さい核種に対しては、光吸収反応を用いた核変換が提案されている。

原子核の光吸収反応断面積には、巨大共鳴と呼ばれるγ線エネルギー約 $80A^{-1/3}$(MeV, Aは核種の質量数)を中心とした幅の広いピーク構造があり、断面積の大部分をこのピークが占めている。したがって、光吸収反応により核変換を行う際は、このピーク領域に対応するエネルギーのγ線を照射するのがもっとも合理的である。

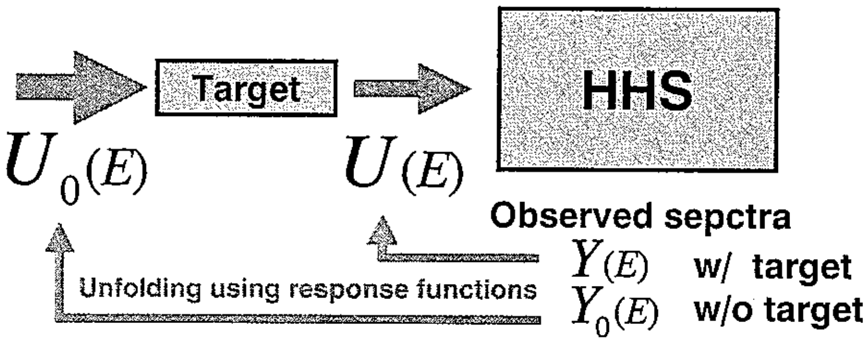
巨大共鳴ピーク構造は、微視的には、様々なエネルギー固有状態間の一粒子-一空孔励起に対応したピークがあるエネルギー領域に集中して出来ていると理解されており、巨大なピークとして観測されるのは十分な測定分解能が無いためであると考えられている。したがって、高分解能検出器を用いて巨大共鳴ピークの微細構造を探索し、個々の一粒子-一空孔励起に対応した微細構造を観測することが出来れば、断面積の最も集中するピークのエネルギーでγ線照射を行うことにより、より効率的な核反応を行うことが出来る。そこで我々は、高分解能高エネルギーγ線スペクトロメータを建設してレーザー逆コンプトンγ線の透過法による測定に用いることにより、光吸収反応断面積の微細構造の探索を行ってきた。

図1に、γ線透過法による光吸収断面積の測定原理を示す。エネルギーEのγ線に対する光吸収反応の断面積$\sigma_{abs}(E)$は、次式で表される:

$$\sigma_{abs}(E)=(1/\rho\ell)\log(U_0(E)/\ U(E))-\sigma_{atom}(E)$$

ここで、$U_0(E)$及び $U(E)$は、照射ターゲット入射前及び透過後のγ線強度を表す。ρ及びℓは、照射ターゲットの密度及び長さを表す。$\sigma_{atom}(E)$は、エネルギーE のγ線の原子による吸収断面積である。しかしながら、吸収断面積を決定するために必要な $U_0(E)$及び $U(E)$を直接測定することは不可能である。というのは、検出器にエネルギーE のγ線が入射した場合、検出器に付与されるエネルギー(応答)が E未満である確率も有限だからである。したがって、我々は $U_0(E)$及び $U(E)$が検出器の応答関数によって畳み込まれてできるγ線スペクトル $Y_0(E)$及び $Y(E)$を観測することになる。このため、光吸収断面積を決定するためには、検出器の応答関数を精密に決定し、それを用いて、観測したスペクトル $Y_0(E)$及び $Y(E)$から $U_0(E)$及び $U(E)$を求める操作、所謂アンフォールディングが必要となる。

現在の技術では、十分に細いエネルギー幅でγ線ビームを作り出すことは不可能であるため、検出器の応答関数を実験的に決定することは出来ない。そこで、近年発達が著しい放射線(γ線及び電子・陽電子)と物質との相互作用のシミュレーションコードを用いて、HHS検出器の実験体系を精密に組み込んだモンテカルロシミュレーションコードを作成し、γ線エネルギー5~25MeVの範囲で、エネルギーステップ100keVでHHS検出器の応答関数を整備した。図2に、作成したシミュレーションコードを用いて得られた、エネルギー5, 10, 15, 20及び25MeVのγ線に対するHHS検出器の応答関数を示す。これにより、γ線透過法によるHHS検出器を用いた高エネルギーγ線の超高分解能測定が可能となった。



$$\sigma_{abs}(E) = \frac{1}{\rho\ell}\log\left(\frac{U_o(E)}{U(E)}\right) - \sigma_{atom}(E)$$

図1: γ線透過法による光吸収断面積の測定の原理。光吸収断面積は、ターゲットにより吸収されたγ線強度 $U(E)$と吸収なしの強度 $U_0(E)$との比較から求まる。しかし、検出器を用いて観測できるのは、検出器の応答関数で畳み込まれたスペクトル $Y_0(E)$及び $Y(E)$である。したがって、検出器の応答関数を正確に決定してアンフォールディングを行うことにより、$Y_0(E)$, $Y(E)$から $U_0(E)$, $U(E)$を求める必要がある。

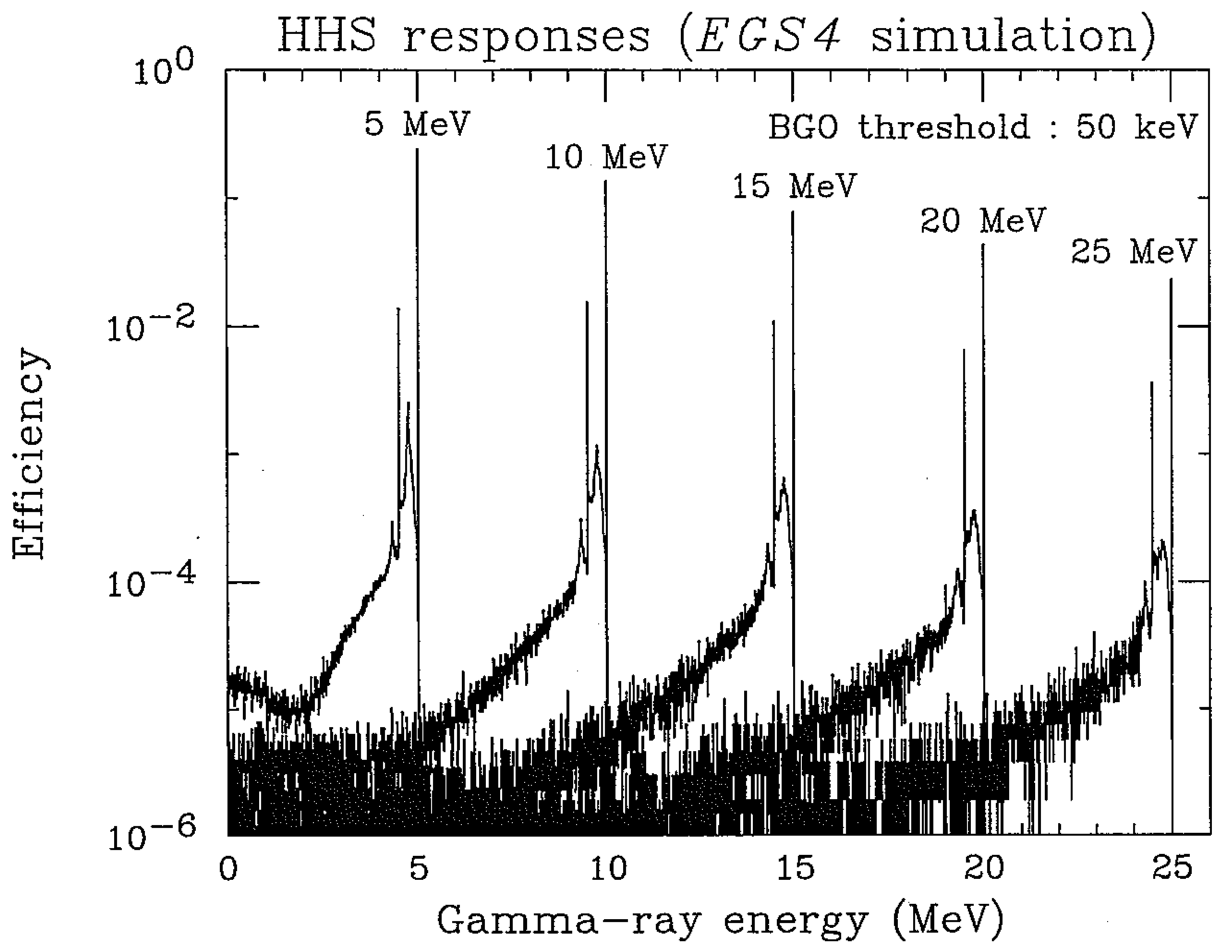


図 2: 開発した HHS 検出器の応答関数シミュレーションコードを用いて得られた HHS 検出器の応答関数(入射 γ 線のエネルギーが 5, 10, 15, 20 及び 25 MeV に対するもの)。

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日本原子力学会「2000 年春の大会(愛媛大)」

$^{137}Cs(n,\gamma)^{138}Cs$ 反応における Cs-138m アイソマー準位生成量の測定
和田浩明、中村詔司、古高和禎、原田秀郎、加藤敏郎、山名元、藤井俊行
日本原子力学会「2000 年春の大会(愛媛大)」

$^{109}Ag(n,\gamma)^{110m}Ag$ 反応の熱中性子吸収断面積及び共鳴積分値の測定
中村詔司、和田浩明、古高和禎、原田秀郎、加藤敏郎
日本原子力学会「2000 年秋の大会(青森大)」

$^{166m}Ho(n,\gamma)^{167}Ho$ 反応の実効熱中性子吸収断面積の測定
原田秀郎、和田浩明、中村詔司、古高和禎、加藤敏郎
日本原子力学会「2000 年秋の大会(青森大)」

(5) ポスター発表(第一著者)

Construction of a γ−γ and β−γ Coincidence Measurement System for Precise Determination of Nuclear Data
K. Furutaka, S. Nakamura, H. Harada, T. Katoh
"*Proceedings of the 1998 Symposium on Nuclear Data*, Nov. 19-20, 1998, JAERI, Tokai, Japan", 181-185
N. Yamano and T. Fukahori (Eds.), JAERI-Conf 99-002, (INDC(JPN)-182/U).

A β−γ Coincidence Measurement System for Precise Determination of g-ray Emission Probabilities of Short-Lived FP Nuclides
K. Furutaka, S. Nakamura, H. Harada, T. Katoh
"*Proceedings of the 1999 Symposium on Nuclear Data*, Nov. 18-19, 1999, JAERI, Tokai, Japan", 160-165
N. Yamano and T. Fukahori (Eds.), JAERI-Conf 2000-005, (INDC(JPN)-185/U).

(6) ポスター発表(共著)

Measurements of Thermal Neutron Capture Cross Sections for Some FP Nuclides
S. Nakamura, K. Furutaka, H. Harada, T. Katoh
"*Proceedings of the 1998 Symposium on Nuclear Data*, Nov. 19-20, 1998, JAERI, Tokai, Japan", 176-180
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第２章 研究の詳細

2.1 Construction of a γ–γ and β–γ Coincidence Measurement System for Precise Determination of Nuclear Data

Construction of a γ–γ and β–γ Coincidence Measurement System for Precise Determination of Nuclear Data

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A γ–γ and β–γ coincidence measurement system was constructed for the precise determination of nuclear data, such as thermal neutron capture cross sections and γ-ray emission probabilities. The validity of the system was tested by a γ–γ coincidence measurement with a ^{60}Co standard source.

1 Introduction

It is of fundamental importance for the nuclear transmutation study of radioactive waste to obtain precise value of nuclear data, such as thermal neutron capture cross sections, $\sigma_{n\gamma}$. From this point of view, we have performed a series of experiments to determine precisely thermal neutron capture cross sections of the FP nuclides by the activation method.

To determine $\sigma_{n\gamma}$ precisely in the conventional activation method, in which γ rays emitted from an activated sample are measured using only one detector, accurate data for γ-ray emission probabilities I_γ are required. For some nuclides, $\sigma_{n\gamma}$ can be determined more precisely if the I_γ of their capture products are obtained with better accuracy.

For the precise determination of $\sigma_{n\gamma}$ and I_γ, a γ–γ and β-γ coincidence measurement system was constructed. In this system, the data is accumulated in list format with singles trigger condition, i.e. singles as well as coincidence data are taken simultaneously. This feature makes dead times of singles and coincidence measurements being canceled out in deducing the activity, so cross sections can be obtained precisely.

In the next section, the feature of the system is described. Section 3 describes an experiment in which γ–γ coincidence measurement was done using standard source, to test the validity of the system. Details of the analysis are also presented in this section, including corrections for sum coincidence and angular correlation. Finally, section 4 summarizes the present work.

2 Feature of the system

A schematic diagram of the system is shown in Figure 1. It consists of two detectors and a fast data acquisition system. Gamma rays are measured by a large Ge detector (relative efficiency 90% of 3"×3" NaI). In β–γ measurements, a thin (0.5–2mm^t) plastic scintillator is used for β detection.

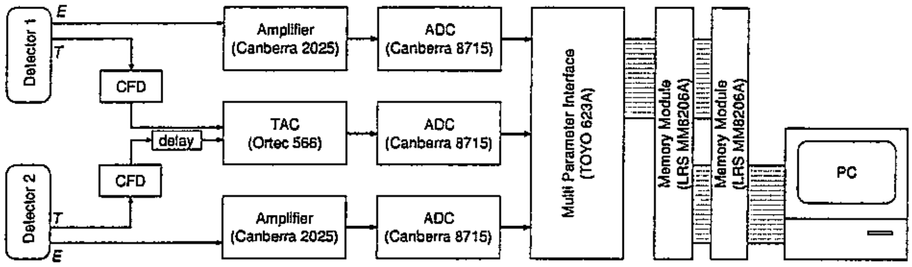

Figure 1: Schematic diagram of the system

The data acquisition system consists of two series of amplifier(Canberra2025)–Fast ADC(Canberra 8715) and a timing circuitry (TAC(ORTEC 566)–ADC(Canberra 8715)) connected to a multi–parameter interface(TOYO 623A). The list data gathered by the interface are temporarily accumulated in one of the two cascade–connected large capacity (64k words) CAMAC memory modules. The data in a memory module are transferred to a PC (NEC PC-9821 Xa16) when the module is not in use, and stored in hard disk of the PC, so the data is accumulated fast and efficiently. The data are accumulated when there's at least one ADC with valid datum. Coincidence events are extracted from the list data in off–line analysis. In γ–ray measurement in singles mode, photo–peak counts N_{i,γ_j} of a γ ray γ_j in a detector i can be expressed as the following:

$$N_{i,\gamma_j} = A \times I_{\gamma_j}\, \epsilon_{i,\gamma_j}\, R_i \times T, \qquad (1)$$

where A is activity, I_{γ_j} emission probability of γ_j, ϵ_{i,γ_j} peak efficiency of γ_j in the detector i, T the measurement time, and R_i the ratio of live–time to elapsed time for the measurement. In γ–γ coincidence measurement, the yield is described by the following equation:

$$N_{1,\gamma_1\,2,\gamma_2} = A \times I_{\gamma_1}\, \epsilon_{1,\gamma_1} \times I_{\gamma_2}\, \epsilon_{2,\gamma_2} \times R_C \times T, \qquad (2)$$

where R_C is live–time ratio for coincidence measurement. According to the above relations, the activity A reads

$$A = \frac{1}{T}\,\frac{N_{1,\gamma_1}\, N_{2,\gamma_2}}{N_{1,\gamma_1\,2,\gamma_2}}\,\frac{R_1\, R_2}{R_C}. \qquad (3)$$

In this system, $R_C = R_1\, R_2$, and

$$A = \frac{1}{T}\,\frac{N_{1,\gamma_1}\, N_{2,\gamma_2}}{N_{1,\gamma_1\,2,\gamma_2}}, \qquad (4)$$

i.e. the dead times in singles and coincidence measurements are canceled out.

3 Validity test of the system

To test the validity of the system, a γ–γ coincidence measurement was performed using a ^{60}Co standard source with known activity. Gamma rays emitted from the source were measured with the two Ge detectors. The detectors were placed face-to-face, and the distance between the source and the front surfaces of the detectors were 20cm. Counting rate for a detector was ~ 2k cps. A total of 1.6×10^7 events were accumulated within about an hour.

Shown in Figure 2 are projection spectra of the energy of the two detectors and TAC data. The two γ rays (1173 and 1333 keV) from the source are clearly seen. Also seen in the figures is 1461 keV peak, which is originated from ^{40}K in the room background. From these projection spectra, yields of the 1173(1333)keV γ ray in Ge-1(Ge-0) was

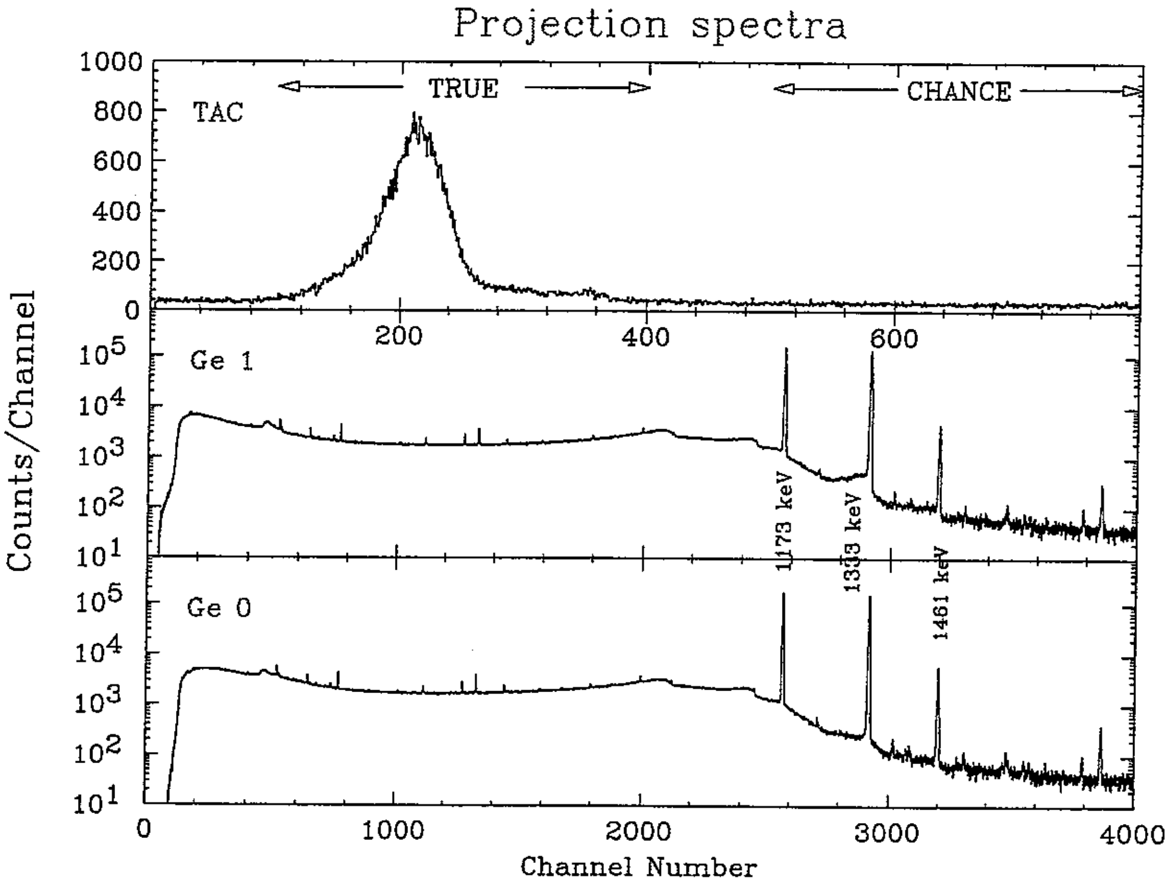

Figure 2: Projection spectra.

obtained.

To extract γ-γ coincidence events, gates were applied to TAC peak ('TRUE' in Figure 2) and Ge-1 1173 keV peak ('G_P' in Figure 3). Spectrum thus obtained is shown in Figure 4.

One of the advantages of list-form data acquisition is that it enables various corrections such as chance coincidence and background contributions, without relying on empirical formula. To estimate contributions from chance coincidence and background component in the coincidence events, separate gates were applied, and the resultant yields were subtracted from the number of coincidence events. Chance coincidence

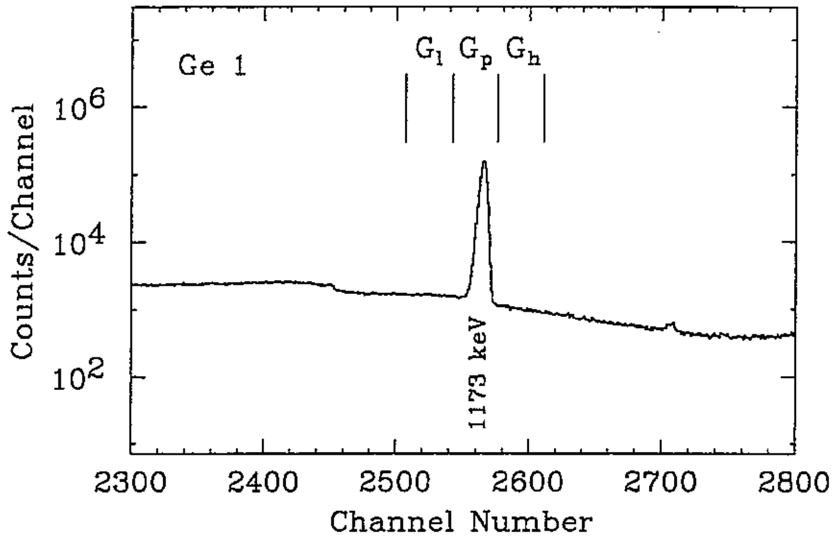

Figure 3: The gates applied to the Ge–1 data.

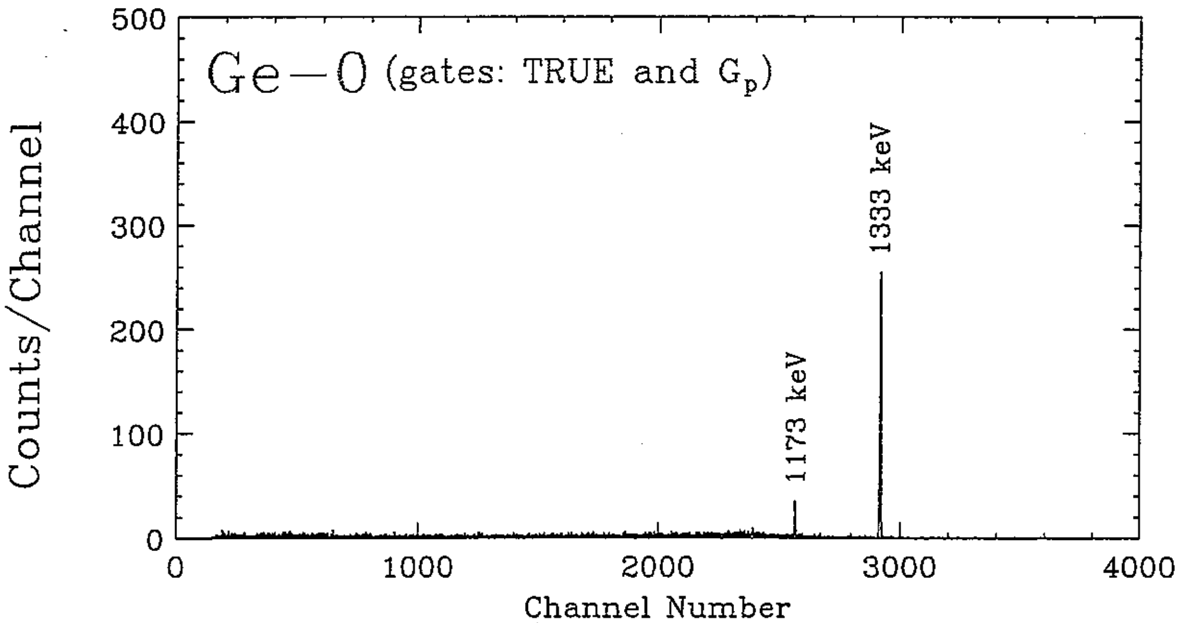

Figure 4: Spectrum obtained by gating Ge–1 1173 keV peak and TAC peak.

contribution was estimated by gating 'G_p' and off-peak portion of TAC data. For background estimation, two gates with the same width were applied on both sides of the peak ('G_l' and 'G_h' in Figure 3), and an average of the resultant counts was regarded as the background.

In addition to the corrections described above, corrections for angular correlation and sum coincidences were made. The correction of coincidence counts, $N_{1,\gamma_1 2,\gamma_2}$, for the angular correlation was done using the following well-known angular correlation function of the two γ rays,

$$W(\theta) = 1 + 0.102 \times P_2(\cos\theta) + 0.0091 \times P_4(\cos\theta), \tag{5}$$

where θ is the opening angle between the two γ rays. The correction factor amounted to 1.112. In sum coincidence events, the two cascade γ rays are detected in the same detector, and therefore singles photo-peak counts are reduced. The correction factors, F_{SC}, of the sum coincidence were calculated for the two cascading γ rays to be

$$F_{SC,1173} = (1.0 - \epsilon_{T,1333} \times f_W)^{-1} \tag{6}$$

and

$$F_{SC,1333} = \left(1.0 - \frac{I_{\gamma,1173}}{I_{\gamma,1333}} \times \epsilon_{T,1173} \times f_W\right)^{-1}, \tag{7}$$

where $\epsilon_{T,1173}(\epsilon_{T,1333})$ is the total detection efficiency of 1173(1333) keV γ ray, $I_{\gamma,1173}(I_{\gamma,1333})$ emission probability of the 1173(1333) keV γ ray, and f_W is the average value of angular correlation function over the detector solid angle weighted by the γ-ray attenuation probability in the Ge crystal. f_W was calculated using the relation (5) above. The factors for the present setup are $F_{SC,1173} = 1.00517$ and $F_{SC,1333} = 1.00531$.

Taking into account all of the above corrections, the activity of the source was obtained as 135.4 ± 5.8 kBq, which agreed to the catalog value with decay correction, 132.5 ± 2.5 kBq. Thus, validity of the system was confirmed in the γ–γ coincidence experiment. The error of the obtained value was mainly (99.6%) originated from the statistical error of the number of coincidence events, and can be reduced with long–time measurements.

4 Conclusion

For precise determination of source activities and γ-ray emission probabilities, a γ–γ and β–γ coincidence measurement system was constructed. Using this system, in which the data are accumulated in essentially singles mode, the dead times for singles and coincidence measurements are canceled out, and the activity can be determined accurately.

To check the validity of the system, the activity of the ^{60}Co standard source was measured with the system using γ–γ coincidence method. The obtained value was agreed to the catalog value within the error, and the validity of the system was confirmed.

2.2 A β−γ Coincidence Measurement System for Precise Determination of γ-ray Emission Probabilities of Short-Lived FP Nuclides

A β-γ Coincidence Measurement System for Precise Determination of γ-ray Emission Probabilities of Short-Lived FP Nuclides

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For the precise determination of γ-ray emission probabilities of short-lived FP nuclides, a β-γ coincidence measurement system has been developed, which utilizes a thin plastic scintillation detector as a β-ray detector. To demonstrate the performance of the developed system, it was applied to the measurement of absolute γ-ray emission probability I_γ of short-lived nuclide, ^{28}Al ($T_{1/2} = 2.24$ (min)), whose I_γ is well known.

1 Introduction

For the nuclear transmutation study of radioactive waste, it is of fundamental importance to obtain precise nuclear data, such as thermal neutron capture cross sections, σ_0. However, some of the available nuclear data are poor in accuracy. In recent years, many works have been extensively done to improve accuracy of such nuclear data using modern radiation detectors and electronics, usually with the activation method[1].

In a conventional activation method, a sample is irradiated with reactor neutrons and γ rays emitted are measured, and the cross sections are deduced from the γ-ray yields. In the calculation, absolute γ-ray emission probabilities I_γ are used. Therefore, precision of I_γ is one of the major factors which determine the precision of the final result, and it is essential that I_γ is precisely determined.

For nuclides with their half lives longer than several minutes, a $4\pi\,\beta$-γ coincidence method has been successfully applied. However, in this method, a radioactive sample has to be set inside a $4\pi\,\beta$-ray gas flow proportional counter so as to attain β-ray detection efficiency to be close to unity, and it takes a couple of minutes to prepare a sample and to set up a β-ray counting system. It is, therefore, difficult with the method to determine absolute γ-ray emission probabilities of short-lived nuclides, whose half lives are considerably shorter than few minutes.

For precise determination of γ-ray emission probabilities of short-lived nuclides, a β-γ coincidence measurement system has been developed, which utilizes a plastic scintillation counter as a β detector and a fast data acquisition system. In this system, energy and timing information are accumulated in event-by-event mode. By using a plastic scintillator for β-ray detection instead of a $4\pi\,\beta$-ray gas flow proportional counter, the time required for the preparation of the radiation detection equipments is greatly reduced, because there is no need of putting a sample inside a detector and setting up a gas-flow β-ray counting system. It is also essential for efficient measurement of radiation from short-lived nuclei to use a fast data processing system which operates at high counting rates. In addition to these, it is inevitable in such high counting condition to determine dead times precisely to extract true coincidence events unambiguously.

2 The β-γ coincidence System

A schematic diagram of the β-γ coincidence system is shown in Figure 1. The system consists of a Ge γ-ray detector, a plastic scintillation β-ray detector and a fast data acquisition system.

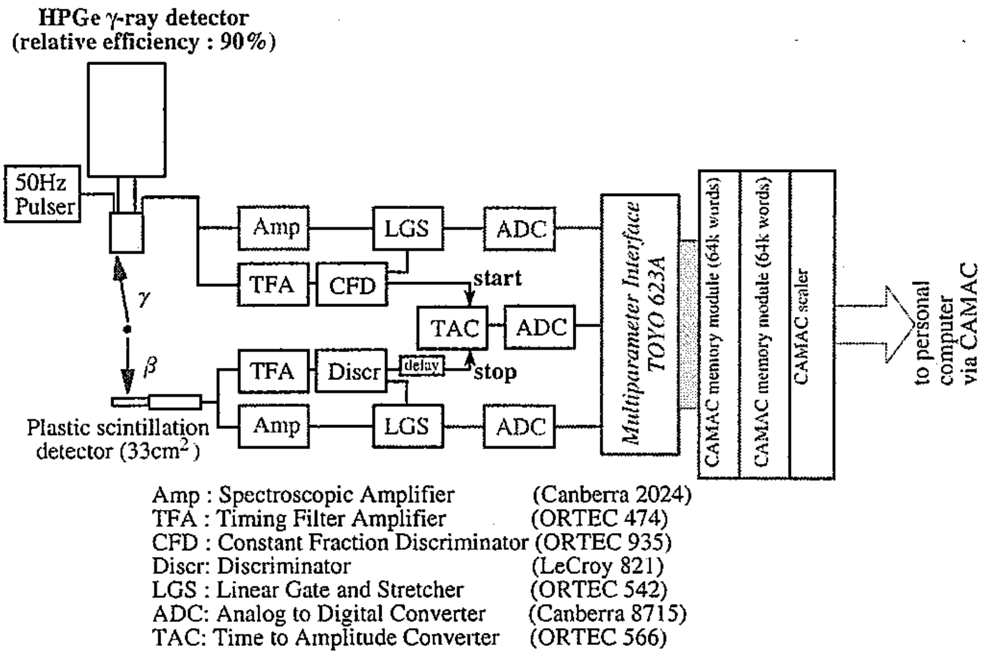

Figure 1: A schematic diagram of the β-γ coincidence system.

A large volume Ge detector whose relative detection efficiency is 90% of that of 7.6 cm×7.6 cm NaI is employed to measure γ rays from short-lived nuclei efficiently. For β-ray detection, a thin and small plastic scintillation detector is used, which is 4 mm in thickness and has an area of 33 cm^2. By using a plastic scintillator as a β-ray detector, the time required for preparation of the irradiated sample and the detection equipments is greatly reduced, and one can efficiently measure radiations from short-lived nuclei. In principle, it is applicable to the measurement of I_γ of a nuclide whose half life is the order of a second.

The energy signal of each detector is fed into an amplifier and then converted by a fast analog-to-digital converter (ADC, Canberra 8715). The timing signal is fed into a discriminator circuit and timing information is extracted. Time difference between the two signals are recorded using a time-to-amplitude converter (TAC) and an ADC. For an efficient measurement of radiations from short-lived nuclei, one should perform a measurement at a high counting rate so as to improve a statistical accuracy. In such a measurement, the TAC information is inevitable to eliminate accidental coincidence events and to extract true number of coincidences.

The data converted by ADCs are processed by a multi-parameter interface module (TOYO 623A) to compose a list of event data. The data are recorded in event-by-event mode to distinguish true coincidence events and to eliminate spurious events such as that caused by accidental coincidence. To reduce dead times in the course of the data acquisition, the data are temporarily accumulated in one of the two memory modules with large capacity (LeCroy MM8206A), which are cyclically connected to operate as a ring buffer, and are transferred to a personal computer for storage when the other memory module is active.

The system is operated in singles trigger condition: when at least one of the two detectors detects a radiation, data of both detectors are stored. The coincidence events are extracted in later off-line analysis.

3 An application to the determination of I_γ in ^{28}Al

To demonstrate the system for an actual I_γ determination of a short-lived nuclide, an experiment was done for 1779 keV γ ray of ^{28}Al. Decay scheme of ^{28}Al is shown in Figure 2. The ^{28}Al decays into ^{28}Si with a half life of 2.24 minutes. In the decay, only one γ ray (1779 keV) is emitted, and the I_γ is known to be unity[2]. In this case, I_γ is expressed as

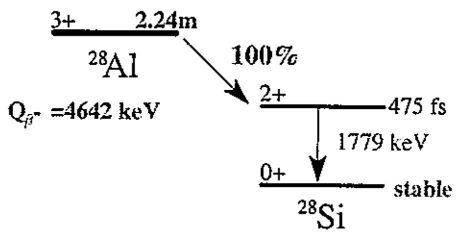

Figure 2: Decay scheme of ^{28}Al.

$$I_\gamma = \frac{1}{\epsilon_\gamma}\frac{n_c}{n_\beta}, \tag{1}$$

where ϵ_γ is detection efficiency of the γ ray, n_β is counting rate of β detector, and n_c is counting rate of β-γ coincidence event.

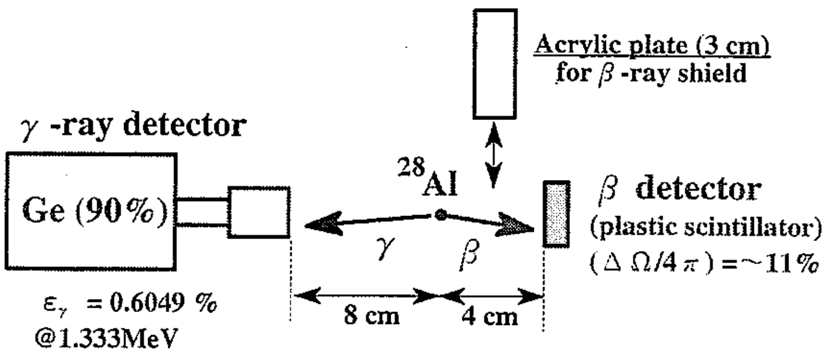

Figure 3: Experimental setup of the I_γ measurement for 1779 keV γ ray in ^{28}Al.

The setup of the experiment is shown in Figure 3. Natural Al foils of 6.76 mg/cm^2 in thickness and 99.0% in chemical purity were irradiated in Rotating Specimen Rack of the research reactor in Rikkyo University. Activity of about 1 MBq was produced by one minute of irradiation, and after cooling of about six minutes, β- and γ-rays were measured for ten minutes. The data were saved every 20 seconds. Measurements were also done with a β-ray shield placed between the irradiated sample and the β detector in order to estimate contribution of γ rays to the β detector. An acrylic plate of 3 cm thickness was used as the β-ray shield.

Shown in upper half of Figure 4 is a typical γ-ray spectrum. As can be seen in the figure, no remarkable γ ray is observed other than 1779 keV γ ray in ^{28}Al. Depicted in lower half of the figure is a decay curve of the 1779 keV γ ray after a correction for dead times. Drawn in the figure in a solid line is a result of fitting the data by a function of the time, t,

$$A e^{-\frac{ln(2)}{\tau} t} + C, \quad (2)$$

where A, τ and C are fitting parameters. The τ means a half life. By averaging the values of τ obtained in all runs, a half life was obtained as $T_{1/2} = 2.248 \pm 0.018$ (min), which agrees with that reported in ref.[2] within the errors, and therefore the γ ray was assigned to be originated from ^{28}Al.

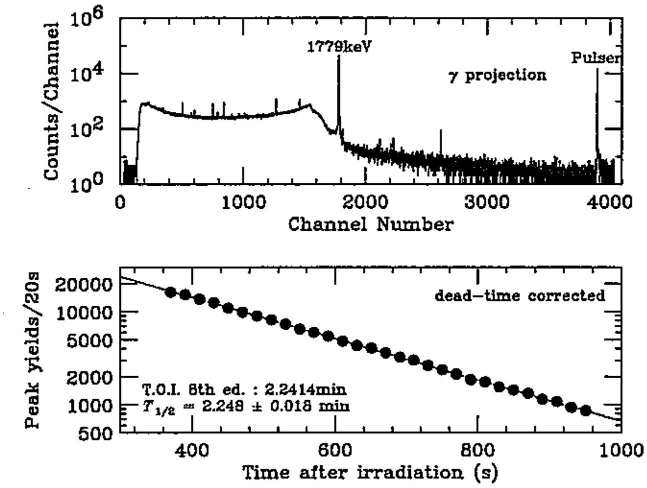

Figure 4: (Upper) : A γ-ray singles spectrum observed in a run. (Lower) : A decay curve of the 1779 keV γ ray in ^{28}Al in the same run, after dead-time correction.

The histogram shown in Figure 5 is a singles β-ray spectrum in a run without the β-ray shield, after subtraction of data in runs with the β shield. A β-ray spectrum obtained by imposing a gate on the 1779 keV γ-ray peak region is also plotted in the figure, which is normalized to the singles one. The singles data deviates from the one gated by 1779 keV γ ray only below about 30 channel, which suggests that there are some events which were not caused by β rays emitted from ^{28}Al. Therefore, lower boundary of summing was varied and the influence to the result was examined when extracting the number of β rays.

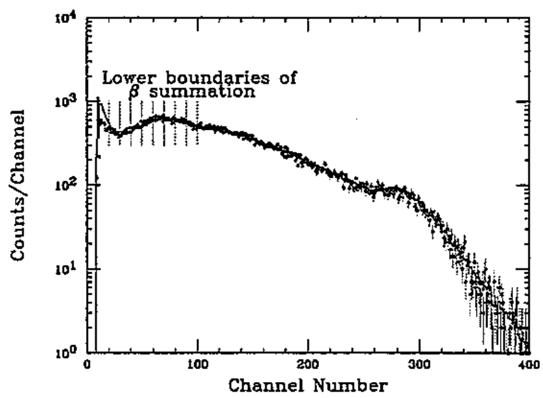

Figure 5: A singles (histogram) and a 1779 keV γ-ray gated (dots) β-ray spectrum in a run.

The number of β rays coincident with the 1779 keV γ ray was deduced by imposing several gates; Γ_P, Γ_H and Γ_L were applied on the γ ray data, while gates T_P and T_O on the TAC data, as shown in Figure 6. Let $n(\Gamma_i, T_j)$ be the number of β counting rate obtained by applying the gates Γ_i and $T_j (i = P, H, L, j = P, O)$ on the γ-ray and the TAC data, respectively. The true β coincidence counting rate n_c is obtained using the following relation,

$$\begin{aligned} n_c = n(\Gamma_P, T_P) \quad &- \quad R \times n(\Gamma_P, T_O) \\ &- \quad R' \left(\frac{n(\Gamma_L, T_P) + n(\Gamma_H, T_P)}{2} - R \frac{n(\Gamma_L, T_O) + n(\Gamma_H, T_O)}{2} \right) \end{aligned} \quad (3)$$

where R represents the ratio of width of T_P to that of T_O, and R' the ratio of width of Γ_P to that of Γ_L.

Shown in Figure 7 are decay curves of β singles (n_β) and coincidence (n_c) counting rates above the 20 channel of β-summing threshold, after the subtraction of data with β-ray shield and the dead-time correction. Dashed lines in the figure represent the result of fitting the data by a function of the same form as in (2), with C and τ fixed at 0 and 2.24 minutes, respectively. A slight deviation is observed in the singles data after about 800 seconds, which seems to be caused by the remaining long life backgrounds. Therefore, the data were fitted with the function (2) and component with half life of 2.24 (min) was extracted. In the fit, τ was fixed at 2.24 minutes and both A and C were varied as free parameters.

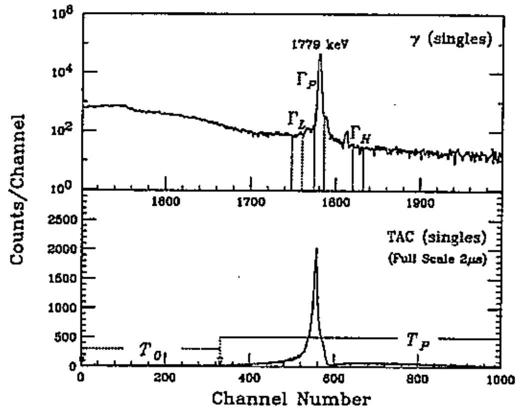

Figure 6: Gates imposed on the γ-ray and TAC data to extract the true number of coincidences.

From the n_β and n_c obtained as above, I_γ of the 1779 keV γ ray was calculated using the relation (1) for each lower limit of β summation. The γ-ray detection efficiency at 1779 keV was determined using ^{60}Co, ^{137}Cs and ^{152}Eu standard sources. Precision of the efficiency was about 2%. The preliminary values of the obtained I_γ are plotted in Figure 8 against the lower limits of β summation. Errors shown in the figure include all the errors except the one of the γ-ray detection efficiency. Above the 30 channel of β summation limit,

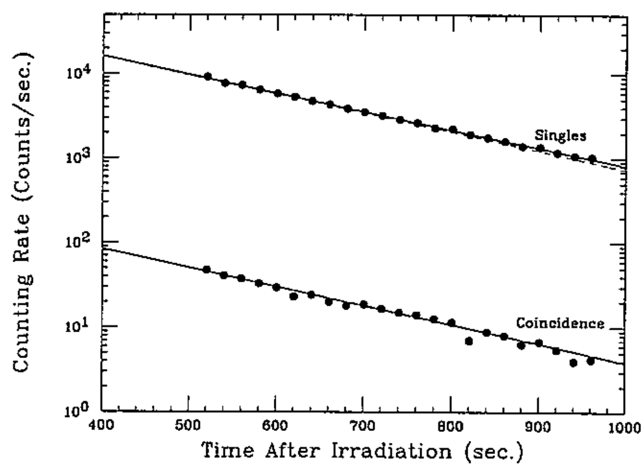

Figure 7: (Points) : Decay curves for β and coincidence channels. (Lines) : Results of the fit with (solid) and without (dashed) a constant term.

the results agree with one another within the errors. The results with errors less than one percent is obtained except an error originated from the determination of ϵ_γ. The obtained results are about two percents smaller than that reported previously [2]. Including the error of the γ-ray detection efficiency, the result agrees with the previous data within the limits of errors.

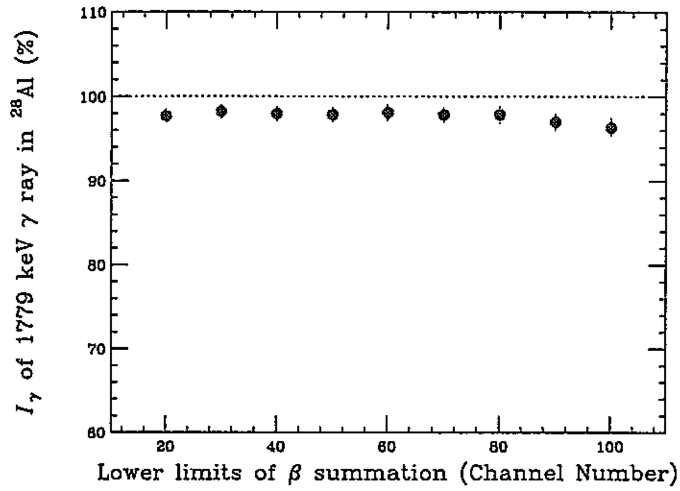

Figure 8: Results obtained in the present experiment for I_γ of 1779 keV γ ray in ^{28}Al.

4 Conclusion

For the precise determination of absolute γ-ray emission probabilities I_γ of short-lived nuclides, a β-γ coincidence measurement system has been developed, which utilizes a thin plastic scintillation detector as a β-ray detector.

The system was applied to the measurement of the absolute γ-ray emission probability of a short-lived nuclide ^{28}Al. The system was demonstrated to have the ability to measure the absolute γ-ray emission probabilities of short-lived nuclides with the precision of $\lesssim$ 2%. By improving accuracy of γ-ray detection efficiency, an absolute γ-ray emission probability will be determined with the total error less than 1%.

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[1] Nakamura, S.: Talk given in this conference.

[2] Firestone, R. B.: "Table of Isotopes", 8th ed., John Wiley & Sons, Inc., New York (1996)

2.3 Evaluation of β−γ coincidence measurement system using plastic scintillation β-ray detector developed for the determination of γ−ray emission probabilities of short-lived nuclides

Evaluation of β-γ coincidence measurement system using plastic scintillation β-ray detector developed for the determination of γ-ray emission probabilities of short-lived nucldes

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Abstract

For precise determination of γ-ray emission probabilities of short-lived nuclides whose half-lives are less than a few minutes, a β-γ coincidence measurement system has been developed which uses a plastic scintillator as a β-ray detector. The system was applied to a measurement of the absolute γ-ray emission probability of 1779 keV γ ray in ^{28}Al ($T_{1/2}$ = 2.24 minutes), whose γ-ray emission probability is well known. The uncertainty of the measured γ-ray emission probability was smaller than 2 %. Performance of the β-γ coincidence system has been demonstrated for a short-lived nuclide, and the details of the corrections has been analyzed.

KEYWORDS:

absolute γ-ray emission probability, short-lived nuclides, β-γ coincidence, plastic scintillator, Germanium detector, ^{28}Al, dead-time correction, high counting rates

和文抄録:

寿命が数分以下の短寿命核種のγ線放出率を精密に測定するために、β線検出器にプラスチックシンチレータを用いたβ-γ同時計測システムを開発した。開発したシステムを用いて、γ線放出率が既知の^{28}Al(半減期2.24分)の1779 keVγ線の放出率測定をおこなった。γ線放出率を誤差2%以下で決定することができ、同システムが短寿命核種のγ線放出率測定に有効であることが実証された。施した補正の詳細について検討をおこなった。

I. INTRODUCTION

An absolute emission probability of a γ ray, I_γ, is one of a fundamental quantity that characterizes the nuclide, and is often used in deducing amount of a nuclide produced in a nuclear reaction. To determine a thermal neutron capture cross section of a nuclide, for example, γ rays emitted after β decays of the capture products are measured and the cross section is deduced from the γ-ray yield. The I_γ is used to deduce the cross section from the obtained γ-ray yields. Therefore, precision and accuracy of the cross section are dependent on the precision and accuracy of I_γ, and it is essential that I_γ is precisely determined.

An absolute γ-ray emission probability is usually determined by β-γ coincidence measurement. For nuclides with half-lives longer than several minutes, a $4\pi\,\beta$-γ coincidence method has been used for determination of I_γ[(1)]. In a conventional $4\pi\,\beta$-γ coincidence method, a radioactive sample is set inside a $4\pi\,\beta$-ray gas proportional counter so as to achieve β-ray detection efficiency close to unity, and it takes a couple of minutes to prepare the sample and to set up the β-ray counting system. For the method to be applicable to a short-lived nuclide, lots of effort has been put into shortening the time required for replacement of gas and a sample. One of the solutions is a use of a multi-anode $4\pi\,\beta$ counter with a slide-type source-mount[(2)], which is operated with CF_4 gas at atmospheric pressure[(3)].

To determine I_γ of a short-lived nuclide, one must measure both β rays and γ rays in a short period before its radioactivity decays out. At the same time, to improve statistical accuracy, one should observe as many decay events as possible. This means that the measurement should be done at high counting rates. Use of a detector with slow timing characteristics in such a condition increases loss of counting. This not only deteriorates the statistical accuracy but also introduces a large systematic error in correcting the loss. Therefore, fast radiation detectors with good timing property should be used in conjunction with a fast data acquisition system which operates well at high counting rates.

By increasing counting rates, the number of accidental coincidence events also increases. At the same time, proportion of dead times becomes larger with the counting rate. These effects lead to a wrong result, and should be corrected. Because the proportions of the accidental coincidence and dead times are dependent on the counting rates of the detectors, and the counting rates are rapidly decreasing in a measurement of radiations from a short-lived nuclide, the corrections should be made for every short interval compared to the half-life.

To determine absolute γ-ray emission probabilities of short-lived nuclides, a β-γ coincidence system has been developed, which uses a plastic scintillation detector as a β-ray detector, a high purity Ge detector as a γ-ray detector, and a fast data acquisition system. By using a plastic scintillator for β-ray detection, whose signal has a rise time and a decay time of the order of nanoseconds, β rays can be measured at higher counting rate than a gas proportional counter. In addition to this, distance between a radioactive sample and a γ-ray detector can be reduced by using a thin scintillator, and one can improve statistical accuracy in the γ-ray yield. In the developed system, energy and timing information are accumulated in event-by-event mode to enable reliable corrections of accidental coincidence and dead times. To enable corrections that are dependent on the counting rates, the developed system stores data into files at each preset interval of a measurement. The system equips data buffers with large capacity to operate well at high counting rate and to minimize dead times in the course of the data acquisition.

The purpose of this paper is to describe the performance of the developed β-γ coincidence system. To demonstrate the ability of the system, the I_γ of short-lived nuclei, ^{28}Al ($T_{1/2}$ = 2.24 (min)), has been measured. This paper is organized as follows: In Section II, details of the β-γ coincidence system are presented. In Section III, the measurement of I_γ of 1779 keV γ ray in ^{28}Al are described. Details of the data analysis are also given in the Section III. Section IV is for discussions of the obtained result and of the data analysis. Conclusions are addressed in Section V.

II. THE β-γ COINCIDENCE MEASUREMENT SYSTEM

A schematic diagram of the β-γ coincidence system is shown in **Fig. 1**. The system consists of a high purity Ge solid-state γ-ray detector, a plastic scintillation β-ray detector, and a fast data acquisition system.

A large volume Ge detector is employed whose relative detection efficiency is 90 % of that of 7.6 cm$\times$7.6 cm NaI detector, to efficiently measure γ rays from short-lived nuclei. It has a built-in transistor-reset type pre-amplifier that enables high counting-rate measurements.

For β-ray detection, a thin plastic scintillator (Nuclear Enterprises, Inc., NE102A) is used. The scintillator is of trapezoidal shape (4.6 and 7.5 cm in lengths of the parallel sides, 5.0 cm in height and 4 mm^t in thickness), and is optically coupled to a metal-package type photomultiplier (PMT, Hamamatsu Photonics K.K., R5600U+E5780) through a light guide. The output signals from the divider of the PMT are directly fed into the processing circuitry. The detector has a rise time of signals of about 3 ns. By using a plastic scintillator as a β-ray detector, β rays can be measured at higher counting rates than that at which the gas proportional counter is able to be operated, and one can efficiently measure radiations from the short-lived nuclei.

The energy and timing information of the radiations are recorded in event-by-event mode. Acquisition of data in event-by-event mode is inevitable to extract the true coincidence events in off-line analysis using the timing information.

The energy signal of each detector is fed into a spectroscopic amplifier (Canberra, 2024 and 2025) and then converted in a fast analog-to-digital converter (ADC, Canberra 8715, which converts a datum in (width of the input signal)+0.8μs). The timing signal is fed into a discriminator circuit and timing information is extracted. Time difference between the two timing signals is converted to digital datum using a time-to-amplitude converter (TAC) and an ADC. The data converted by the ADCs are fed to a multi-parameter interface module (TOYO 623A) to generate a list of event data. To reduce dead times in the course of the data acquisition, the list data are at first accumulated in one of the two dual port memory modules with large capacity (LeCroy MM8206A), which are cyclically connected to operate as a ring buffer, and are transferred to a personal computer for storage while the other memory module is active. The accumulated data are partitioned off at each preset interval and saved into separate files on a hard disk of the computer. This enables various corrections which are dependent on counting rates to be done accurately. The system is operated in singles trigger condition: when at least one of the two detectors detects a radiation, data of both detectors are stored. Whether an event is β-γ coincidence event or not is determined in later off-line analysis. This enables reliable elimination of spurious events based on the recorded timing information.

III. AN APPLICATION TO THE DETERMINATION OF I_γ IN ^{28}AL

To demonstrate the applicability of the system to actual I_γ determination of a short-lived nuclide, the I_γ of 1779 keV γ ray in ^{28}Al has been measured. The ^{28}Al decays via β^- transition into ^{28}Si with a half-life of 2.24 min(4). Decay scheme of ^{28}Al is shown in **Fig. 2**. The ^{28}Al decays to a 2^+ state in ^{28}Si, and only one γ ray is emitted during the decay. The I_γ of 1779 keV γ ray decaying from the 2^+ state is well determined to be unity(4). Conversion electrons are not emitted. In this case, the data analysis is simple, and therefore the nuclide is suited to demonstrate the ability of the system for the determination of I_γ.

The singles counting rates of the β rays n_β and γ rays n_γ are expressed by the following relations,

$$n_\beta = A\,\varepsilon_\beta, \tag{1}$$

$$n_\gamma = A\,\varepsilon_\gamma\,I_\gamma, \tag{2}$$

where A denotes the activity of the sample, ε_β detection efficiency of the β ray, and ε_γ peak detection efficiency of the γ ray. In the same way, the β-γ coincidence counting rate n_c is expressed as

$$n_c = A\,\varepsilon_\beta\,\varepsilon_\gamma\,I_\gamma. \tag{3}$$

Angular correlation between β and γ ray is neglected in the above relation, because it is expected only for first and higher order forbidden β transitions[5]. These three relations lead to the following expression of I_γ,

$$I_\gamma = \frac{1}{\varepsilon_\gamma}\frac{n_c}{n_\beta}. \tag{4}$$

1. The experimental setup

The experiment was carried out at The Institute for Atomic Energy of the Rikkyo University. Natural foils of aluminum (99 % in chemical purity) were irradiated for 60 seconds with neutrons in Rotating Specimen Rack of the research reactor TRIGA MK-II at the Institute. Thickness of the foils is 25 μm (6.75 mg/cm^2). Size of the foils are about 5 mm$\times$5 mm. The irradiated sample was placed between the β- and γ-ray detectors as shown in **Fig. 3**. The distances between the sample and the front surfaces of the detectors were 8 cm for γ-ray detector and 4 cm for β detector, respectively.

In the experiment, the shaping times of the amplifiers were set to 0.5 μs and 4 μs for β- and γ-ray channels, respectively. The widths of the output signals from the linear gate modules were all set to 2 μs.

Radiations emitted from a sample were measured for 10 minutes. The data were saved every 20 seconds. Measurements were also done with a β-ray shield placed between the sample and the β detector, to estimate susceptibility of the plastic scintillator to radiation other than β rays. An acrylic resin plate of 3 cm in thickness was used as the β-ray shield. The range of ^{28}Al β rays in an acrylic resin is estimated to be 1.2 cm with a semi-empirical formula[6]. Therefore, the shield stops the β rays completely. A total of 11 measurements were done including 5 runs with the β-ray shield.

As can be seen from the relation (4), the γ-ray peak detection efficiency ε_γ should be accurately calibrated in order to determine I_γ precisely. The ε_γ was calibrated using ^{152}Eu and ^{60}Co standard γ-ray sources. The activity of the ^{60}Co source was accurately determined by a series of γ-γ coincidence measurements. Precision of the activity of the ^{60}Co source is 0.48 %. Precision of the activity of the ^{152}Eu source was so poor as 8.7 %. The ^{152}Eu, however, emits γ rays ranging from 122 keV to 1408 keV, and their relative strengths are well established[7]. Therefore, the ^{152}Eu source is useful in determining γ-ray energy dependence of the detection efficiency. The ε_γ was assumed to be dependent on the γ-ray energy E_γ in the following form: $\varepsilon_\gamma(E_\gamma(\text{MeV})) = a\exp(b\,\log(E_\gamma))$ (%), where a and b are constants. The slope parameter b was determined by measuring the γ rays from the ^{152}Eu standard source positioned in the same place as a ^{28}Al sample. The summed-coincidence effect for cascading γ rays in ^{152}Eu has been corrected. Then the normalization parameter a was determined by measuring the γ rays from the ^{60}Co source. The parameters were determined to be $a = 0.701 \pm 0.001$ and $b = -0.591 \pm 0.007$, and the efficiency at 1779 keV was 0.499 ± 0.002 % for the present setup. The details of the γ-γ coincidence measurements will be presented elsewhere[8].

2. Data analysis and the result

(1). The projection spectra

Shown in **Fig. 4 (a)** is a typical γ-ray projection (singles) spectrum. As can be seen from the figure, no remarkable γ ray other than 1779 keV is observed below about 4 MeV. A decay curve of the γ ray that was obtained for the same run, after dead-time correction described later, is plotted in **Fig. 4 (b)**. A solid line drawn in the figure was obtained by fitting the data with a function of the time, t,

$$N\,\exp(-\frac{\ln 2}{\tau}\,t) + C, \tag{5}$$

where N, τ and C are fitting parameters. The τ means a half-life. By averaging the values of τ obtained in all runs, a half-life was obtained as $T_{1/2} = 2.248 \pm 0.002$ (min) which agrees with that, 2.2414 ± 0.0012 (min), reported in Ref. 4 within the limits of errors. The half-life of ^{28}Al is also reported in Ref. 9 to be 134.93 ± 0.009 (s) (2.2488 ± 0.0002 (min)), with which the present result agrees better. Therefore the γ ray was assigned to be originated from ^{28}Al.

Shown in **Fig. 5** in a solid line is a typical β-ray singles spectrum obtained in a run without the β-ray shield, after subtraction of data obtained in runs with the β-ray shield. Solid circles in the figure represents a β-ray spectrum obtained by imposing a gate on 1779 keV γ-ray peak region. The gated spectrum is normalized to the singles one. The singles spectrum deviates from the gated one only below about 30 channel, which suggests that there are some events which were not caused by β rays emitted from ^{28}Al. Therefore, when counting the number of β rays detected in the plastic scintillator, lower limit of the summation was varied and the influence to the result was examined.

Shown in **Fig. 6** (a) is a TAC spectrum observed in a typical run without the β-ray shield. A peak which originated from β-γ coincidence events is clearly observed in the spectrum. The asymmetry observed in shape of the peak was caused by a difference of the rise time between the Ge and the plastic detector.

(2). Extraction of the true number of coincidence events

In principle, the number of coincidence events (and hence the coincidence counting rate) can be deduced by selecting the events whose γ-ray energy and TAC data are in the corresponding peak regions and by counting the number of gated β particles. The events thus selected, however, include not only accidental coincidence events but also events that are caused by Compton scattering of γ rays with higher energy than that of interest and the energy deposited in the detector happen to be the same as that of the peak (Compton tail coincidence events). If such events are included in the number of coincidence events, a wrong result will be obtained, in particular in a measurement at a high counting rate such as that encountered in a coincidence measurement of radiations from a short-lived nuclei. Formulae for correction of accidental coincidence have been presented by several authors

(Refs. 5, 10, 11). No general formula exists for correction of the Compton tail coincidence events. Therefore, a correction for the Compton tail coincidence events should be done based on the observed spectral shapes. The TAC information is essential in the correction.

To eliminate these spurious coincidence event and extract true coincidence counts, several gates were applied on both the γ-ray data and TAC data, namely $\Gamma_{\rm P}$, $\Gamma_{\rm H}$ and $\Gamma_{\rm L}$ on the γ-ray data and $T_{\rm P}$ and $T_{\rm O}$ on the TAC data, as shown in **Fig. 6 (a)** and **(b)**. The gate $\Gamma_{\rm P}$ selects events with γ-ray data in the γ-ray peak region, whereas $\Gamma_{\rm L}$ and $\Gamma_{\rm H}$ with γ-ray data below and above the peak region, respectively. The $T_{\rm P}$ denotes a gate for selecting events whose TAC data is in the peak region, $T_{\rm O}$ off-peak events which are recorded by accidental coincidence. Let $n'(\Gamma_i, T_j)$ be the number of β counting rate obtained by applying the gates Γ_i and T_j (i=P,H,L, j=P,O) on the γ-ray energy and on the TAC data, respectively. The true β coincidence counting rate $n'_{\rm c}$ is calculated using the following relation,

$$\begin{aligned} n'_{\rm c} = {} & n'(\Gamma_{\rm P}, T_{\rm P}) - R \times n'(\Gamma_{\rm P}, T_{\rm O}) \\ & - R' \times \left(\frac{n'(\Gamma_{\rm L}, T_{\rm P}) + n'(\Gamma_{\rm H}, T_{\rm P})}{2} - R \times \frac{n'(\Gamma_{\rm L}, T_{\rm O}) + n'(\Gamma_{\rm H}, T_{\rm O})}{2} \right), \end{aligned} \tag{6}$$

where R represents the ratio of width of $T_{\rm P}$ to that of $T_{\rm O}$, and R' the ratio of width of $\Gamma_{\rm P}$ to that of $\Gamma_{\rm L}$; width of $\Gamma_{\rm H}$ is taken to be the same as that of $\Gamma_{\rm L}$. The second term in the right hand side of the relation represents correction for accidental events, whereas the third term is correction for Compton tail coincidence events. In **Fig. 7 (a)**, $n'_{\rm c}$ and $n'(\Gamma_{\rm P}, T_{\rm P})$ in a typical run are plotted against the time after irradiation. The ratios $n'(\Gamma_{\rm P}, T_{\rm P})/n'_{\rm c}$ were also plotted in **Fig. 7 (b)**. As can be seen from the figure, at 470 s after irradiation, as much as 14 % of the events in $n'(\Gamma_{\rm P}, T_{\rm P})$ were due to the spurious coincidence events. Also, it can be seen from the figure that the ratio is rapidly changing with time. Therefore, in a measurement of radiations from a short-lived nuclide, correction for the dead times should be done for each short interval in which the decrease in counting rate is not large. Also plotted in the figure are ratios of $n'(\Gamma_{\rm P}, T_{\rm P}) - R\,n'(\Gamma_P, T_{\rm O})$ to n'_c, which represent ratios of the number of coincidences that was corrected only for accidental coincidence to the one that was corrected both for accidental coincidence and Compton tail coincidence. Although the ratios are on the average 0.3 % larger than unity in the present experiment, correction for Compton tail coincidence events will be more important in a measurement in which many numbers of γ rays are emitted from the radioactive nuclide of interest.

(3). Dead-time corrections

Before the relation (4) is used to deduce I_γ, corrections for dead times and γ-ray events in the β detector should be made. Formulae for dead-time corrections of singles and coincidence counting rates have also been discussed in Refs. 5, 10, 11. In the present analysis, a prescription described in Ref. 11 was employed. Let $n'_\beta(l)$ and $n'_\mathrm{c}(l)$ be respectively the observed β and coincidence counting rates above a lower limit of β summation l, n'_β and n'_c the corresponding total observed counting rates. Similarly n'_γ and n_γ represent the observed and true γ-ray counting rates and let τ_β and τ_γ be the dead times of the respective channels which include not only the dead times of the detectors but also dead times of the electronic circuitry. The true β ($n_\beta(l)$) and coincidence ($n_\mathrm{c}(l)$) counting rates above l are calculated using the following relations,

$$n_\beta(l) = D_\mathrm{s}\, n'_\beta(l), \tag{7}$$

$$n_\mathrm{c}(l) = D_\mathrm{c}\, n'_\mathrm{c}(l), \tag{8}$$

$$D_\mathrm{s} = \frac{1}{1 - n'_\beta \tau_\beta}, \tag{9}$$

$$D_\mathrm{c} = \frac{1}{1 - n'_\beta \tau_\beta - n'_\gamma \tau_\gamma + n'_\mathrm{c} \tau_\beta}, \tag{10}$$

assuming $\tau_\beta < \tau_\gamma$.

The dead times τ_β and τ_γ were determined in a separate measurements using the same setup, in which counting rate of each of the detectors was varied using a standard radioactive source and losses of signals of a precision pulse generator was observed. In the measurements it was confirmed that the ratio of the number of recorded signals to that of generated pulses decreased linearly with the apparent counting rate up to a counting rate of 10k cps. The obtained values were $\tau_\beta = 6.38 \pm 0.18$ μs and $\tau_\gamma = 32.56 \pm 0.30$ μs. In **Fig. 8**, the observed counting rates for β and γ ray in a run without β-ray shield, n'_β and n'_γ (a), and the resultant dead-time correction factors for β-ray singles and coincidence, D_s and D_c (b), are plotted as a function of time. It should be noted that, as is seen from the figure, D_c amounted to about 16 % at 470 s after irradiation. This indicates an importance of the dead-time correction in a high counting-rate measurement. Also note that the dead-time correction factors are rapidly varying functions of the time. This also indicates the need of the correction at short intervals.

After the dead-time corrections, γ-ray events detected in the β counter were subtracted. All of the events observed in runs with the β-ray shield were assumed to be caused by 1779 keV γ rays. Data for runs with the β shield were summed and normalized by a ratio of weight of sample, and then subtracted from the data without the β shield. Attenuation of the 1779 keV γ rays in the β shield was also taken into account, and corrected using atomic photoabsorption cross sections tabulated in Ref. 12.

(4). The results

The open circles and solid circles in **Fig. 9** show decay curves of the β singles ($n_\beta(l)$) and the β-γ coincidence ($n_c(l)$) counting rates with $l = 30$ in a run, respectively, where the dead times were corrected and the backgrounds were subtracted in the manner as described in the previous subsection. Dashed lines in the figure represent the result of fitting the data by a function of the same form as in (5), with C and τ fixed at 0 and 2.248 min, respectively. While no visible deviations are observed in n_c, slight deviations are observed in n_β, which seems to be caused by the remaining long-life backgrounds. Therefore, the data were fitted with the function (5) with τ fixed at 2.248 min and both N and C varied as free parameters, and contributions from components with half-life of 2.248 min were extracted. In the fit, the parameter C was constrained to be positive. The results of the fits were indicated in the figure in solid lines.

Then, the ratios $n_c(l)/n_\beta(l)$ were calculated for each run from the results of the fit, and then averaged over all runs without the β-ray shield. The ratios are plotted in **Fig. 10 (a)**. Errors of the ratios, which include both statistical and systematic ones, are smaller than the size of solid circles. and the magnitude of the errors was as small as 1.0 %.

Finally, I_γ was obtained from ratios $(1/\varepsilon_\gamma)(n_c(l)/n_\beta(l))$. These ratios are plotted against the lower limit of β summation, l, in solid circles in **Fig. 10 (b)**. The results agree with one another within the limits of errors, and essentially independent of l. This is natural, because no conversion electron is radiated and only one γ ray is emitted in the decay of ^{28}Al nucleus. From the ratios, I_γ of 1779 keV γ ray in ^{28}Al was determined as 0.995 ± 0.014. The obtained value agrees with that reported in Ref. 4 within limits of errors. Error of the obtained result is as small as 1.4 %. Therefore, the ability of the system for the determination of an absolute γ-ray emission probability of a short-lived nuclide was successfully demonstrated in the case where only one γ ray is emitted. Also plotted in the figure in open circles are I_γ values obtained from an analysis in which corrections for dead times and γ-ray attenuation in the β-ray shield were not made. The results without the corrections are about 10% smaller than that with the corrections. This fact clearly indicates an importance of the dead-time correction for a measurement at a high counting rate.

IV. DISCUSSION

1. Uncertainty of the obtained results

The errors of the obtained result (0.995 ± 0.014) include:

- statistical error
- error resulting from the assumption that in runs with the β-ray shield all events observed in the β-ray detector were caused by the 1779 keV γ rays (< 0.43 %),
- errors in n_β and n_c that is caused by fixing the parameter τ to 2.248 min (0.56 %),
- error resulting from the β-ray discrimination level l (0.49 %),

- error in the γ-ray detection efficiency resulting from the ambiguity of the calibration source intensity (0.5 %),
- error in the γ-ray detection efficiency resulting from the ambiguity of the source position (0.91 %).

It should be noted that errors of the ratios $n_c(l)/n_\beta(l)$, i.e. total errors except for the ones in detection efficiency of the γ ray and in β-ray discrimination level l, are as small as 1.0 %. This again shows the ability of the system to the determination of I_γ. As much as 64 % of the total errors were resulted from the error in the determination of the detection efficiency of the γ ray. Therefore, if γ-ray detection efficiency is determined more precisely, an absolute γ-ray emission probability is more precisely determined using the present method. This can in part be achieved by reducing ambiguity in the activity of the source.

2. Application to a nuclide with complex decay scheme

In the present paper, an application of the developed β-γ coincidence system to the determination of absolute γ-ray emission probability of 1779 keV γ ray in ^{28}Al is described. In this case, only one γ ray is emitted. Also, no conversion electron is emitted. Therefore, the relations (1)–(4) suffice the analysis.

To apply the system to determine I_γ of nuclides with more complex decay schemes, detection efficiency of the conversion electrons and contribution from more than one β branches have to be taken into account in the analysis as in Ref. 13.

V. SUMMARY AND CONCLUSION

For the precise determination of γ-ray emission probabilities of short-lived nuclides whose half-lives are less than a few minutes, a β-γ coincidence measurement system has been developed which uses a plastic scintillator as a β detector. The system has been applied to a measurement of absolute γ-ray emission probability of 1779 keV γ ray in ^{28}Al, whose γ-ray emission probability is known to be unity. In analyzing the obtained data, corrections were done for dead times, accidental coincidence events, Compton tail coincidence events for each short interval. In addition to these, correction for γ-ray events detected in the β detector was done. An absolute γ-ray emission probability of 1779 keV γ ray in ^{28}Al was obtained as 0.995 ± 0.014. The result agrees with the value reported previously within the limits of errors. The error of the results was as small as 1.4 %. Therefore, the system have an ability to measure the absolute γ-ray emission probabilities of short-lived nuclides. The system can be used to obtain precise and accurate I_γ for many kind of short-lived nuclides, whose I_γ could not be accurately determined by using a conventional coincidence measurement system.

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FIGURE CAPTIONS

Fig. 1. A schematic diagram of the β-γ coincidence system.

Fig. 2. Decay scheme of ^{28}Al.

Fig. 3. Experimental setup of the I_γ measurement for 1779 keV γ ray in ^{28}Al.

Fig. 4. (a) A γ-ray singles spectrum observed in a run. (b) A decay curve of the 1779 keV γ ray in ^{28}Al (squares) in the same run as in (a), after dead-time correction. Errors are smaller than size of the squares. The solid line represents the result of a fit to the data with a function of the form (5).

Fig. 5. A singles (solid line) and a 1779 keV γ-ray gated β-ray spectra (dots) observed in a run without the β-ray shield, after subtraction of data obtained in runs with the β shield.

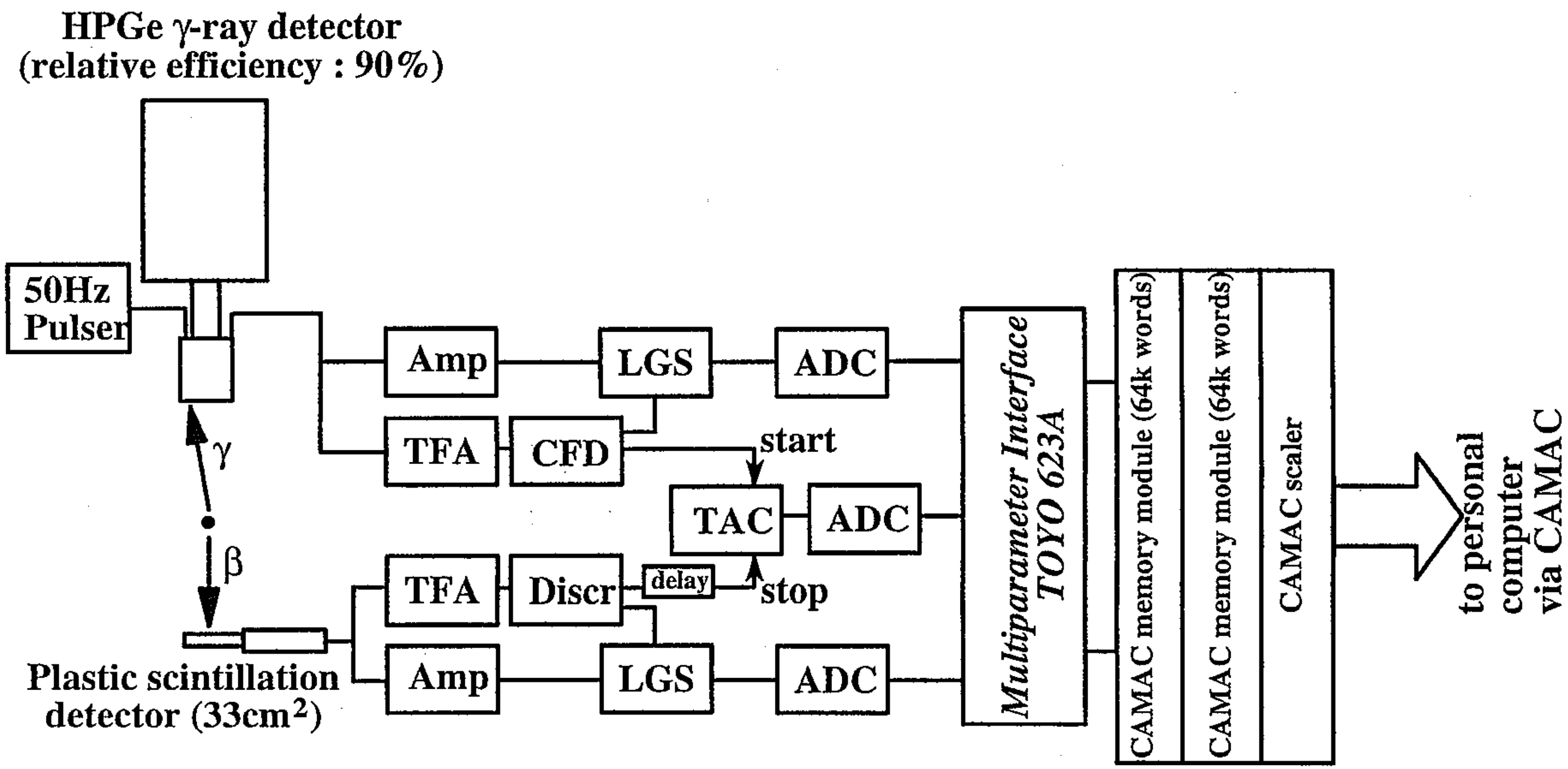
Fig. 6. (a) A typical TAC spectrum and the gates applied to the data. (b) The gates imposed on the γ-ray energy.

Fig. 7. (a) The coincidence counting rate, corrected (open circle) and not corrected (solid circle) for accidental coincidence events. (b) The ratios of the uncorrected ($n'(\Gamma_{\rm P}, T_{\rm P})$) to the corrected ($n'_{\rm c}$) counting rates. Errors are not shown. Open squares represent the ratios of $n'(\Gamma_{\rm P}, T_{\rm P}) - R\,n'(\Gamma_{\rm P}, T_{\rm O})$ to n'_c.

Fig. 8. (a) The observed β- (n'_β, solid circles) and γ-ray (n'_γ, solid squares) singles counting rate. (b) The dead-time correction factors for β-ray singles ($D_{\rm s}$, open circles) and coincidence ($D_{\rm c}$, solid circles) counting rates, as a function of time.

Fig. 9. The decay curves of the true β-ray singles (n_β, open circles) and coincidence ($n_{\rm c}$, solid circles) counting rates. Lines show the results of the fit with (solid) and without (dashed) a constant term. For $n_{\rm c}$, the two lines overlap.

Fig. 10. (a) The ratios n_c/n_β for each β summation lower limit l averaged over all runs without the β-ray shield. (b) The obtained values of γ-ray emission probability of 1779 keV γ ray in ^{28}Al for each lower limit of β summation l (solid circles). The results, which are obtained by an analysis without dead-time corrections and corrections for γ-ray attenuation in the β-ray shield, are also shown in open circles. Errors of the results without the corrections include statistical ones only.



Amp : Spectroscopic Amplifier (Canberra 2024)
TFA : Timing Filter Amplifier (ORTEC 474)
CFD : Constant Fraction Discriminator (ORTEC 935)
Discr: Discriminator (LeCroy 821)
LGS : Linear Gate and Stretcher (ORTEC 542)
ADC: Analog to Digital Converter (Canberra 8715)
TAC: Time to Amplitude Converter (ORTEC 566)

Fig. 1

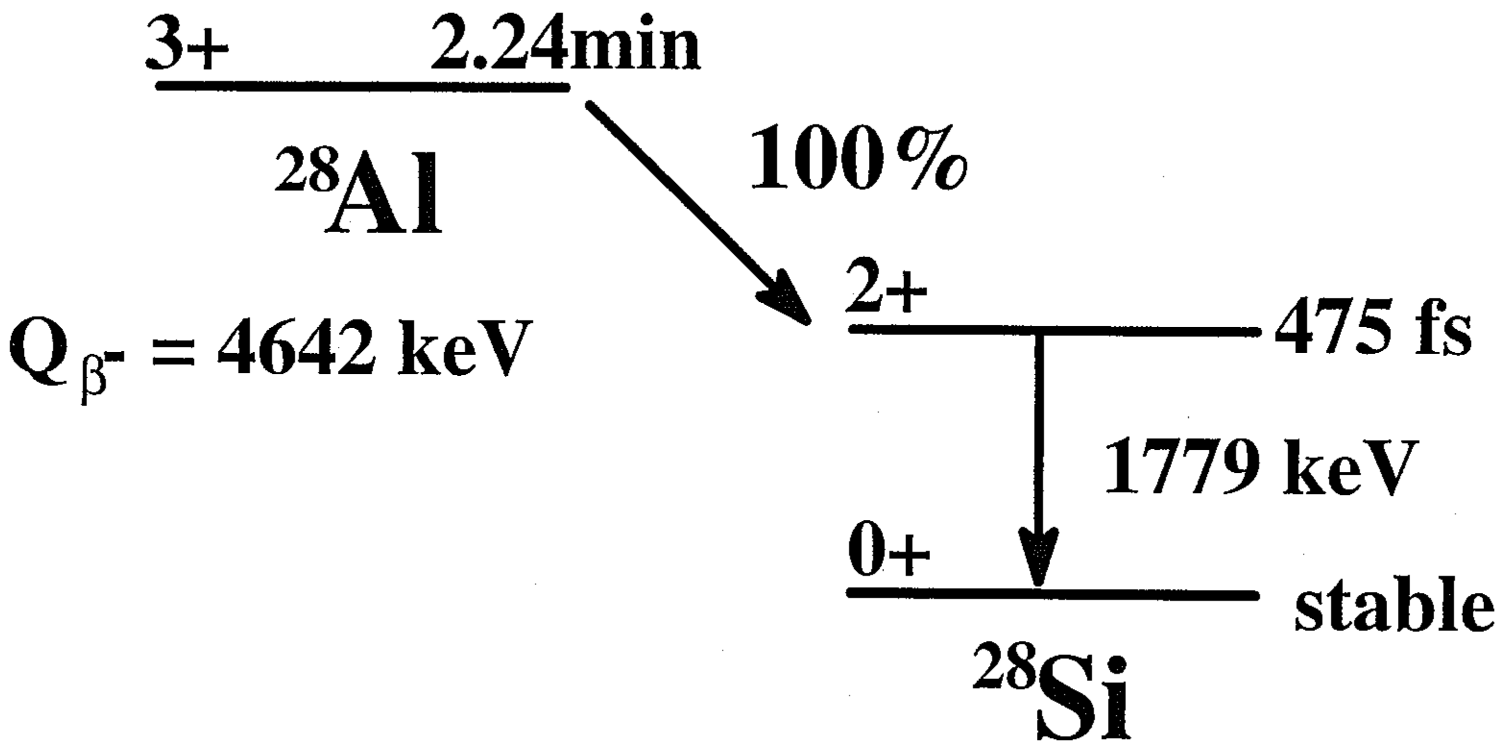


Fig. 2

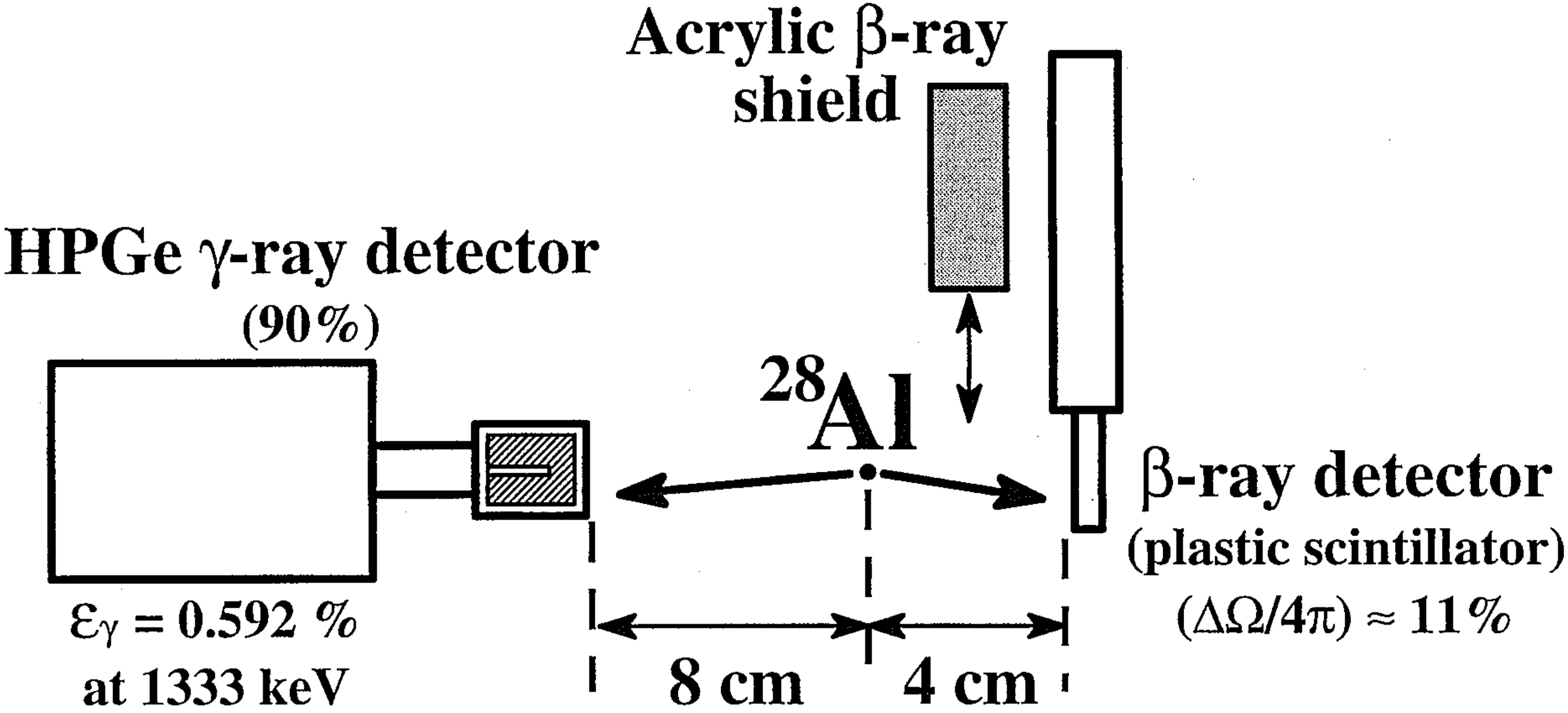
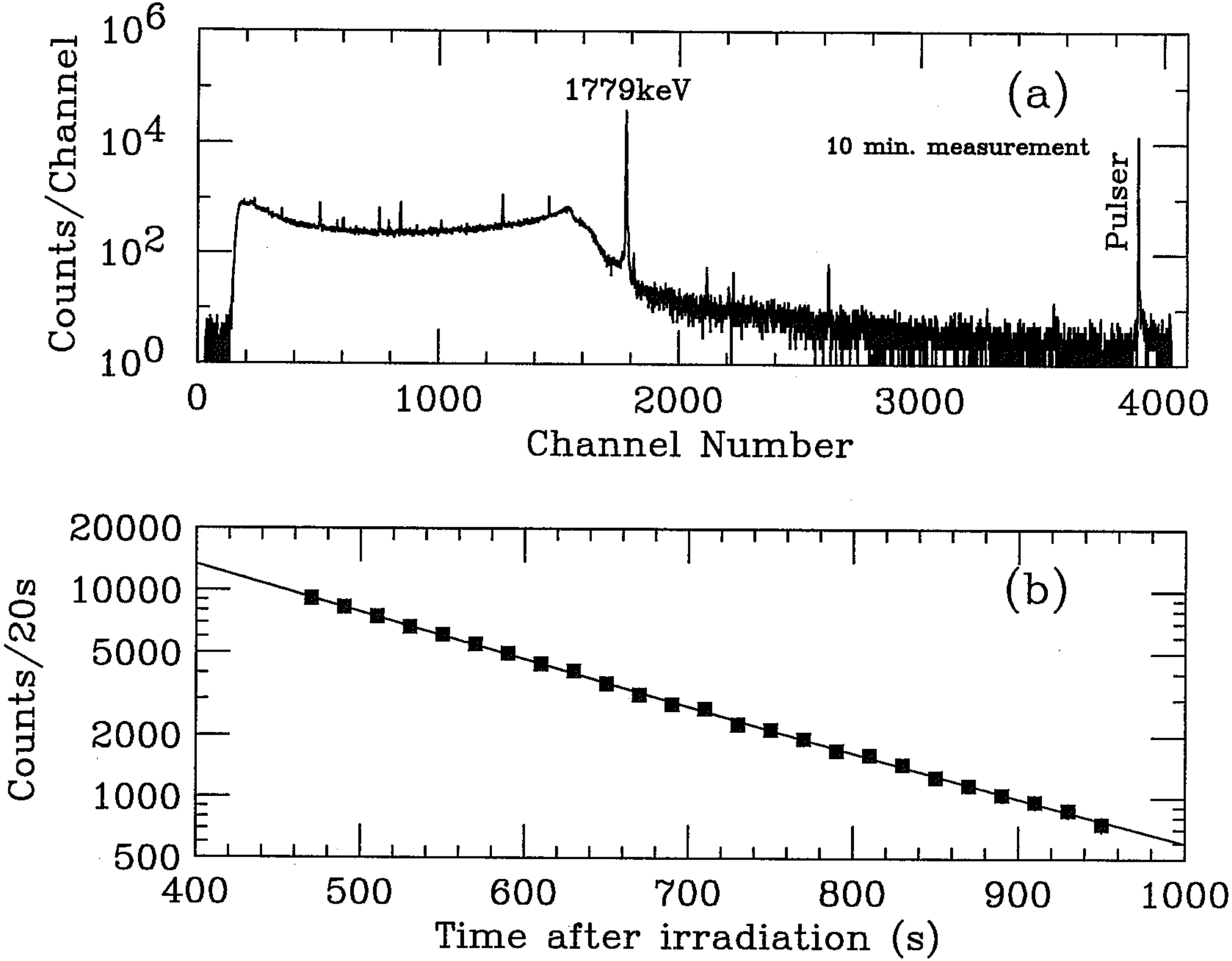

Acrylic β-ray
shield
HPGe γ-ray detector
(90%)
28Al
β-ray detector
(plastic scintillator)
(ΔΩ/4π) ≈ 11%
εγ = 0.592 %
at 1333 keV
8 cm
4 cm

Fig. 3



(a)
1779keV
10 min. measurement
Pulser
Counts/Channel
10^0
10^2
10^4
10^6
0
1000
2000
3000
4000
Channel Number
(b)
Counts/20s
500
1000
2000
5000
10000
20000
400
500
600
700
800
900
1000
Time after irradiation (s)

Fig. 4

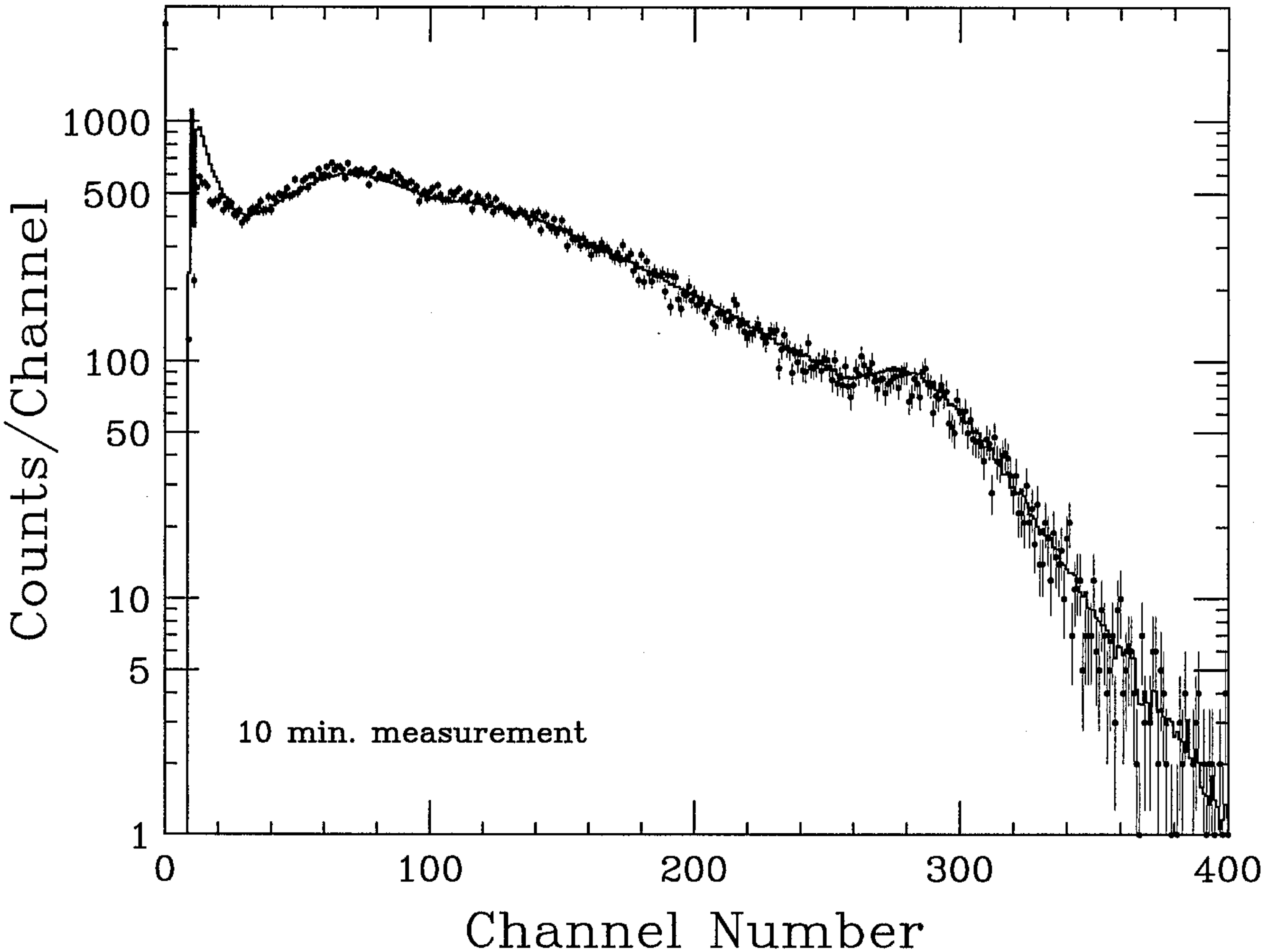


Fig. 5

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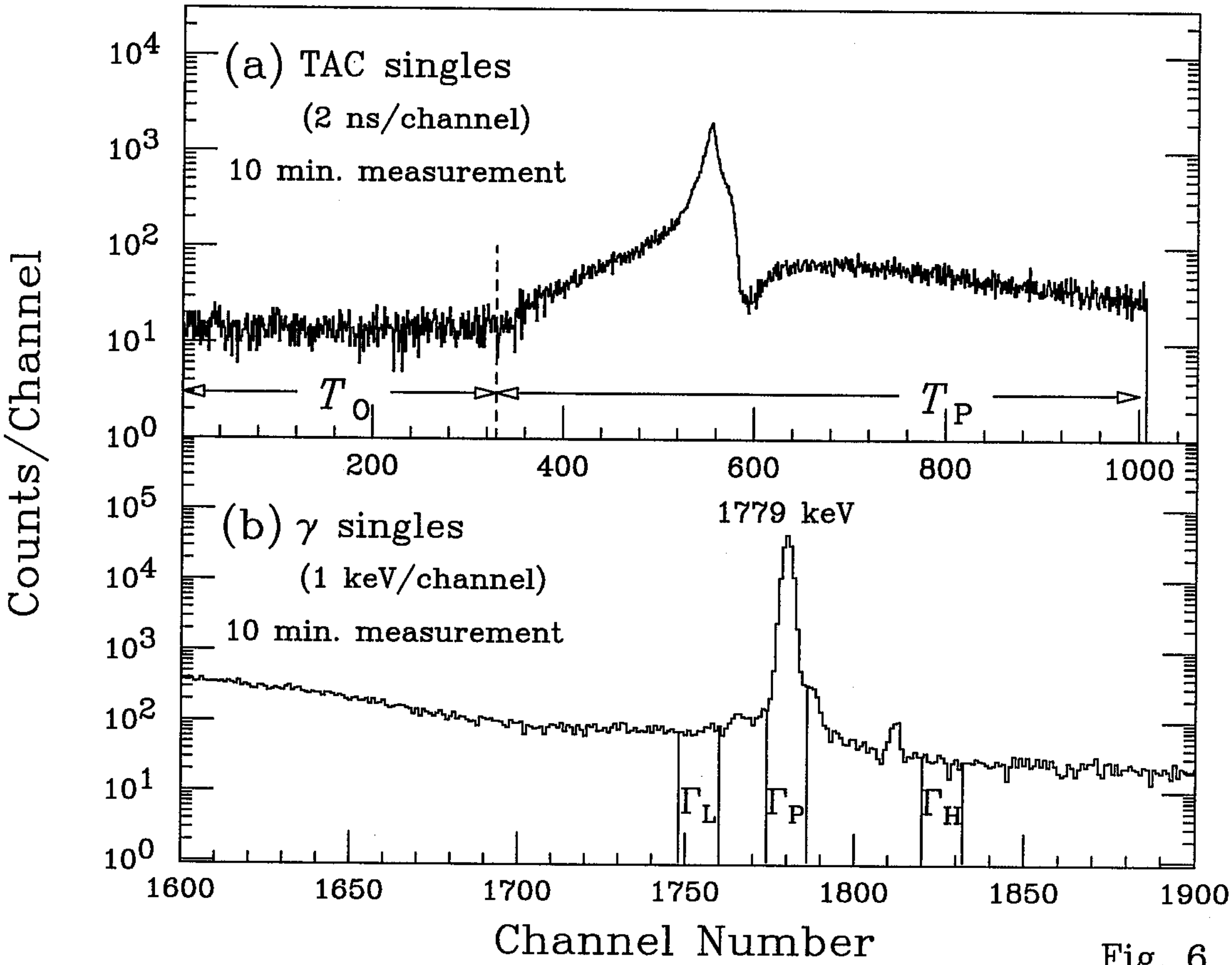

Counts/Channel
(a) TAC singles
(2 ns/channel)
10 min. measurement
T_O
T_P
200
400
600
800
1000
(b) γ singles
(1 keV/channel)
10 min. measurement
1779 keV
Γ_L
Γ_P
Γ_H
1600
1650
1700
1750
1800
1850
1900
Channel Number

Fig. 6

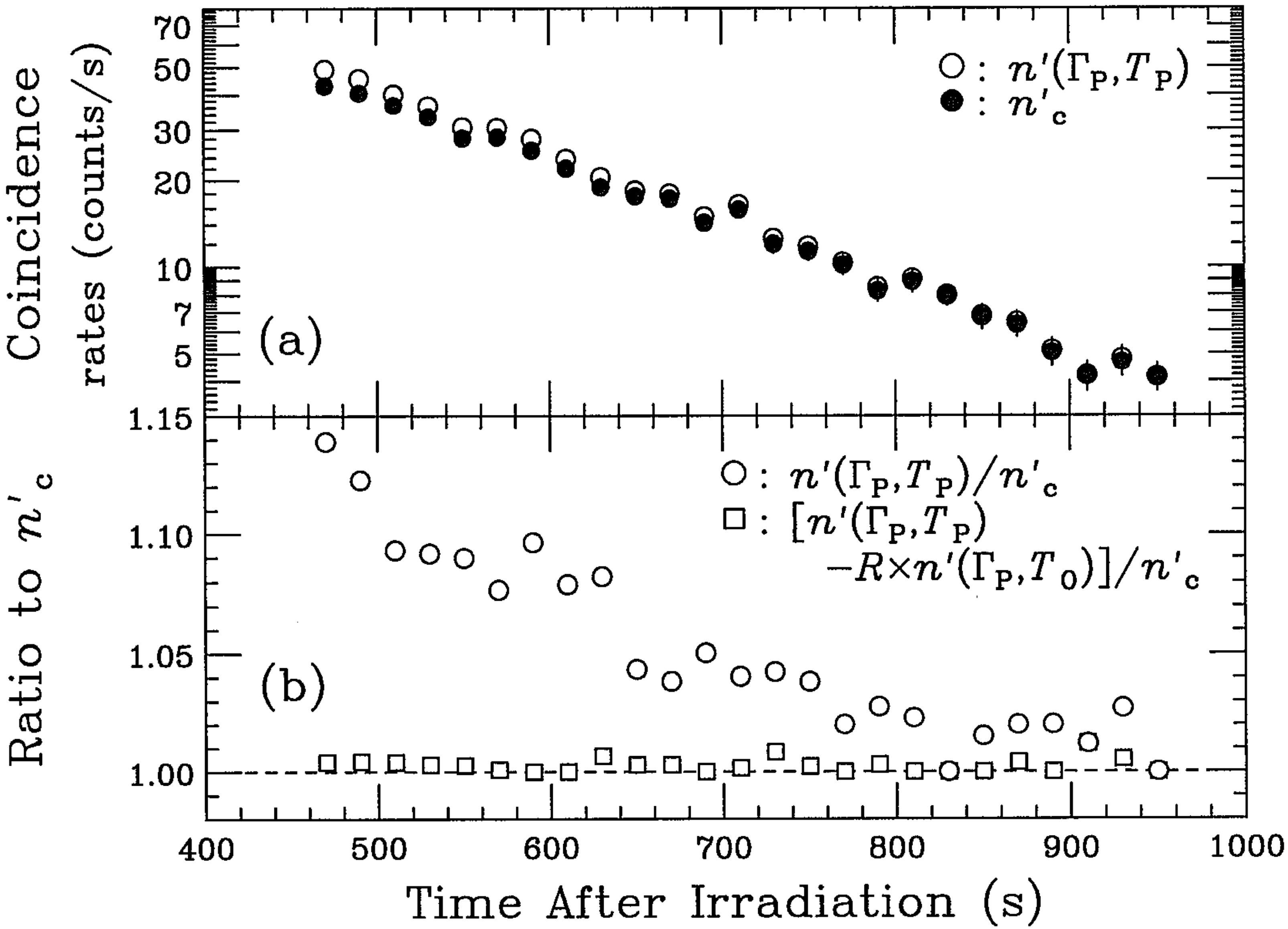

Coincidence rates (counts/s)
70
50
30
20
10
7
5
(a)
○ : $n'(\Gamma_P, T_P)$
● : n'_c
Ratio to n'_c
1.15
1.10
1.05
1.00
(b)
○ : $n'(\Gamma_P, T_P)/n'_c$
□ : $[n'(\Gamma_P, T_P)$
$-R \times n'(\Gamma_P, T_0)]/n'_c$
400
500
600
700
800
900
1000
Time After Irradiation (s)

Fig. 7

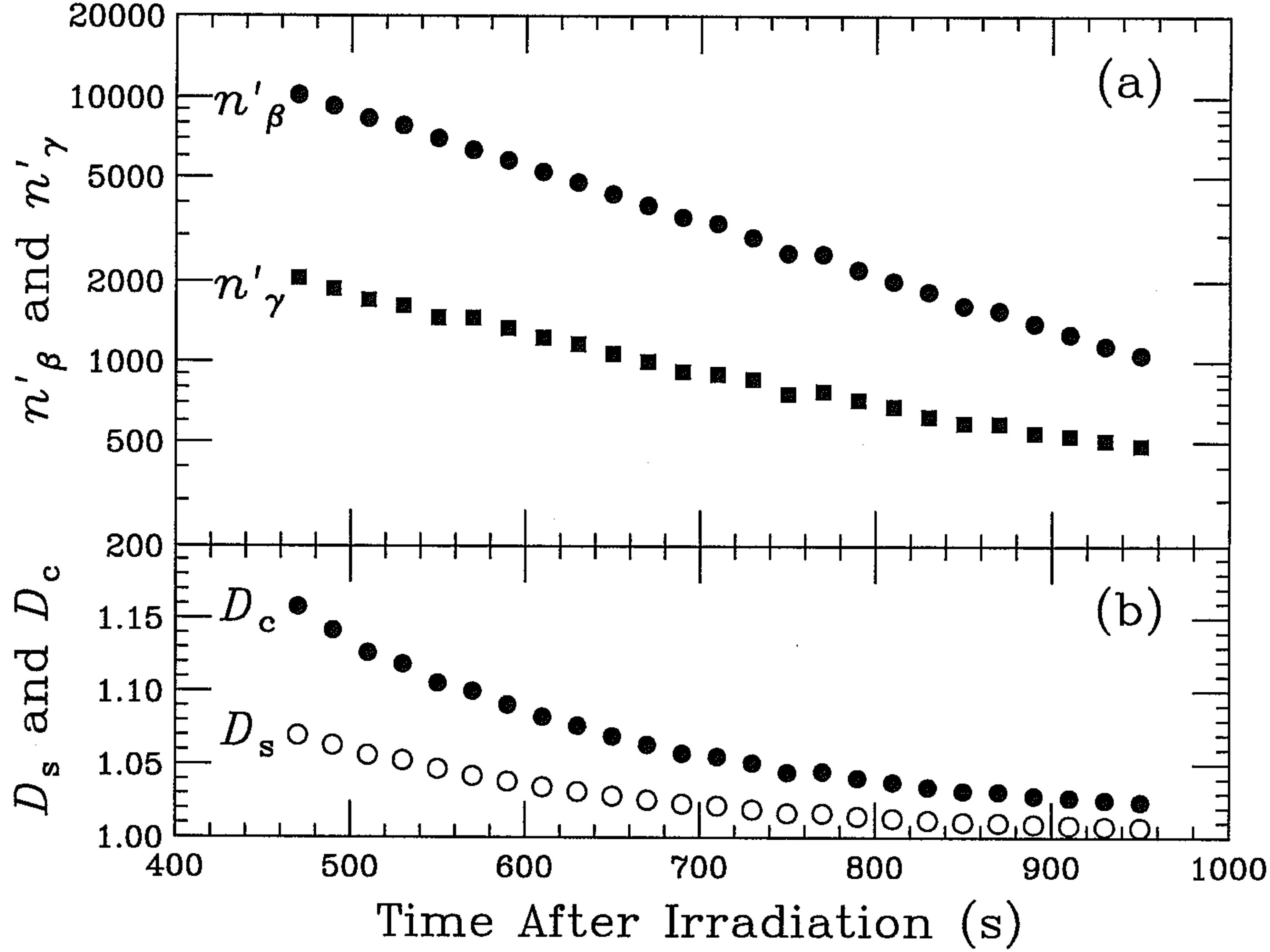

(a)
(b)
n'_β
n'_γ
D_c
D_s
n'_β and n'_γ
D_s and D_c
20000
10000
5000
2000
1000
500
200
1.15
1.10
1.05
1.00
400
500
600
700
800
900
1000
Time After Irradiation (s)

Fig. 8

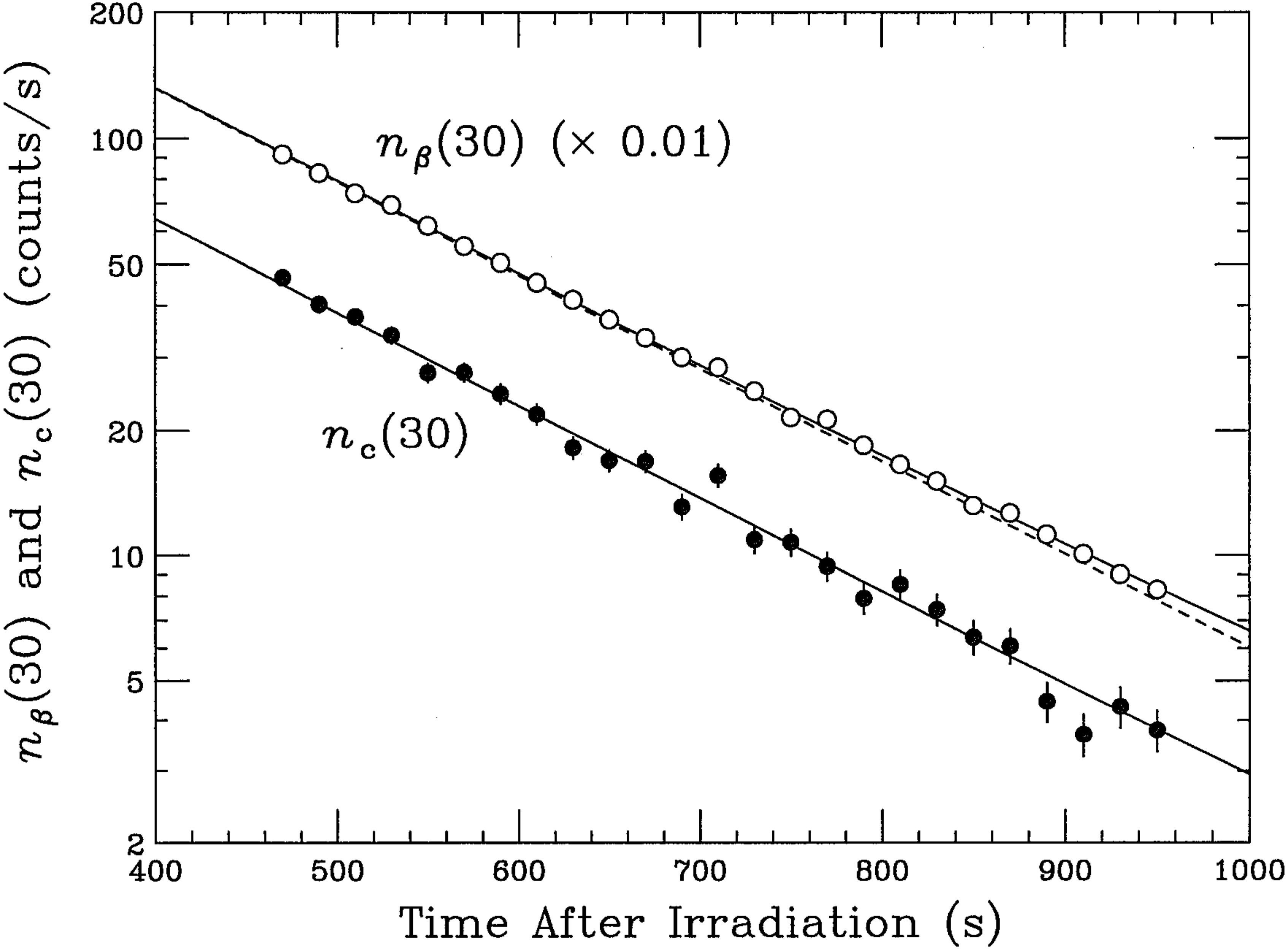

$n_\beta(30)$ ($\times$ 0.01)
$n_c(30)$
$n_\beta(30)$ and $n_c(30)$ (counts/s)
Time After Irradiation (s)
200
100
50
20
10
5
2
400
500
600
700
800
900
1000

Fig. 9

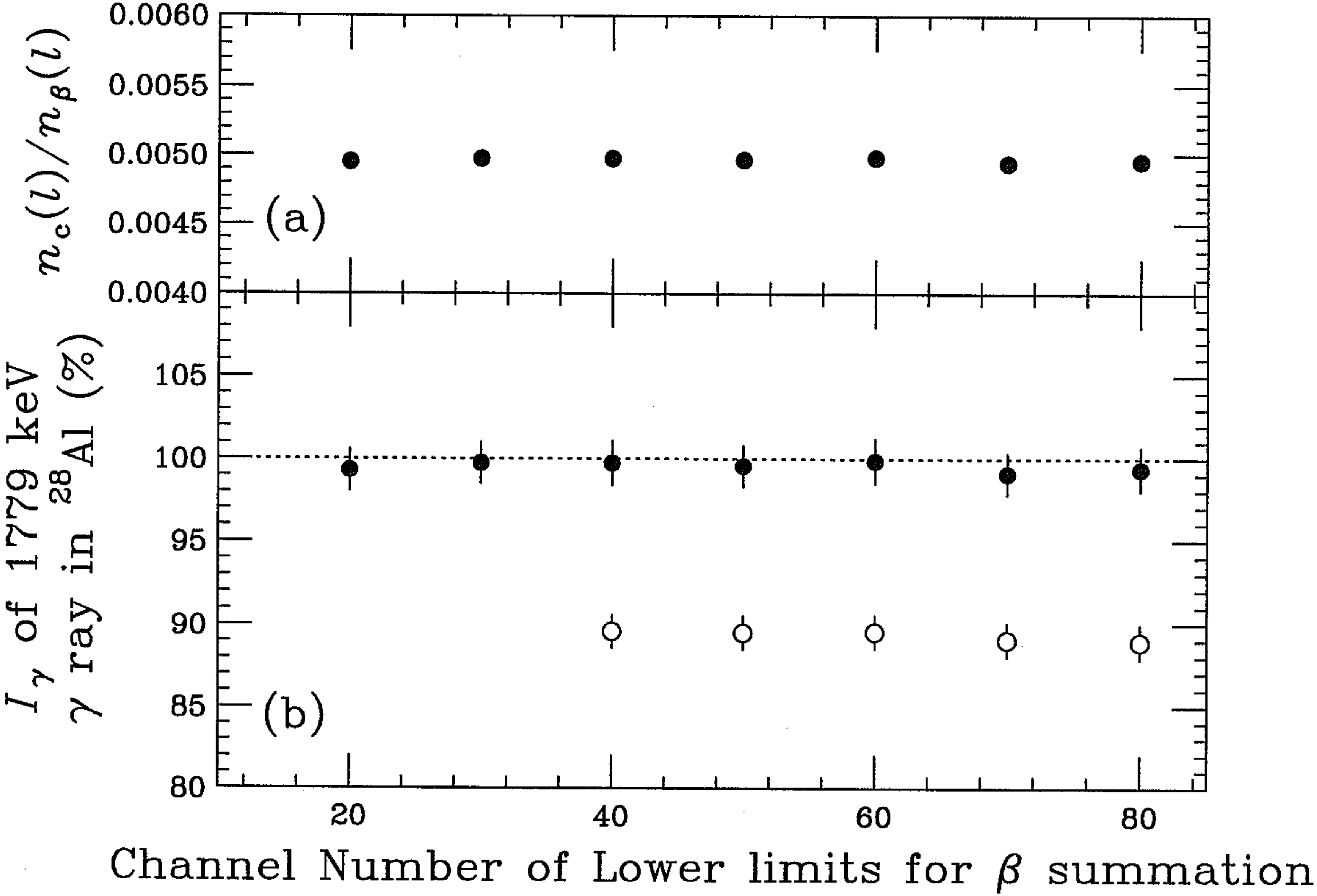

0.0060
0.0055
0.0050
0.0045
0.0040
$n_c(l)/n_\beta(l)$
(a)
105
100
95
90
85
80
I_γ of 1779 keV
γ ray in ^{28}Al (%)
(b)
20
40
60
80
Channel Number of Lower limits for β summation

Fig. 10

2.4 Precise Measurements of γ-ray Emission Probabilities of ^{100}Tc

Precise Measurements of γ-ray Emission Probabilities of ^{100}Tc

JNC

K. Furutaka, S. Nakamura, H. Harada, T. Katoh

KURRI

T. Fujii, H. Yamana

^{99}Tc

▲ Long-Lived Fission Product ($2.1x10^{5}$ y)

▲ Large Fission yield (~6.1%*)

▲ High geochemical mobility

- A candidate for neutron transmutation

Main FP Nuclei

Half - Life	Nucleus
>5000y	^{93}Zr, ^{79}Se, ^{126}Sn, ^{107}Pd, ^{129}I, ^{135}Cs, **^{99}Tc**, (^{14}C)
>100y	^{94}Nb, ^{166m}Ho, ^{158}Tb, ^{108m}Ag
>30y	^{151}Sm, ^{121m}Sn, ^{90}Sr, ^{137}Cs
<30y	^{152}Eu, ^{85}Kr, ^{134}Cs etc.

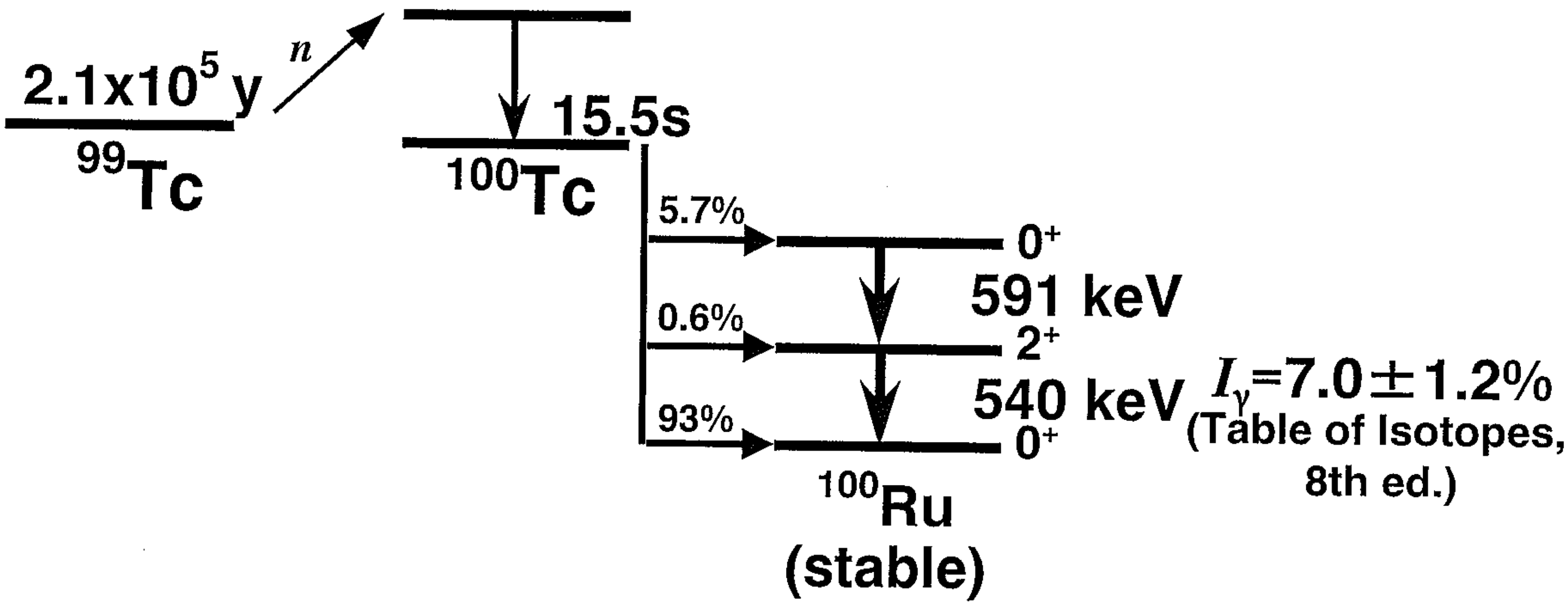
*** For ^{235}U(thermal), from JNDC-V2**

Measurements of thermal neutron capture cross section of ^{100}Tc using an activation method

▲ γ-ray emission probabilities (I_γ) of ^{100}Tc

- Not precise
- Not accurate

Large errors in the resultant cross section



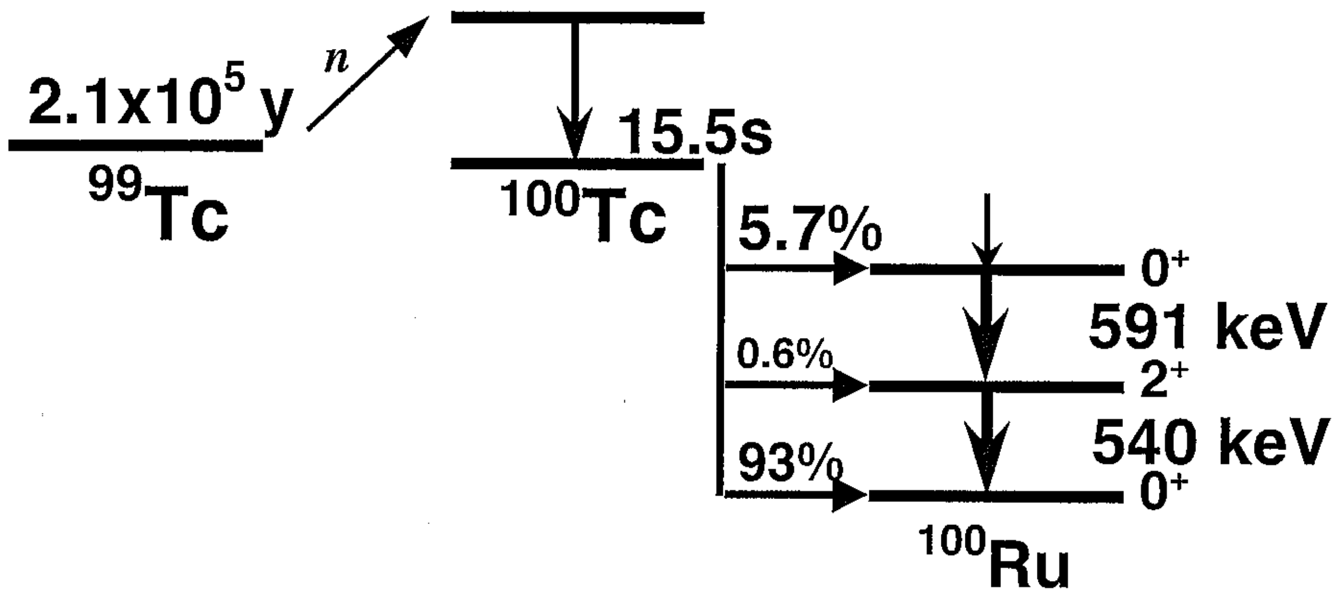
Original data of I_γ for ^{100}Tc

▲ β-γ coincidence experiment

(G. Berzins *et al.* Phys. Rev. 187, 1618 (1969))

- γ : NaI(Tl)
- β : Plastic (not a 4π geometry, ε_β differs for each branch)
- β feedings to excited states other than 1131 keV levels were neglected

$(1/\varepsilon_\gamma)\ n_c(591\text{keV})/n_\beta = 5.7\%$ ⇒ $b_2 = 5.7\%$



Improved β-γ coincidence measurement of I_γ for ^{100}Tc

▲ β-γ coincidence experiment

- γ : HPGe (90%)

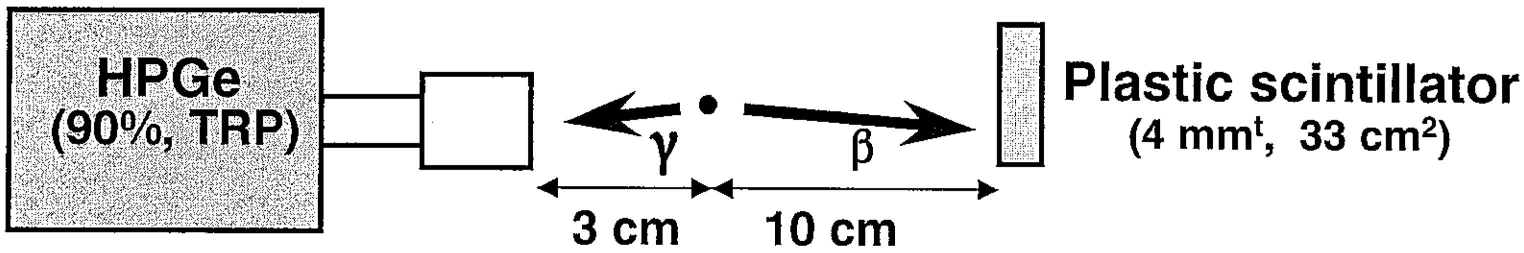
  ⇒ Improvement in accuracies of γ-ray yields

- β : Plastic (not a 4π geometry, ε_β differs for each branch)

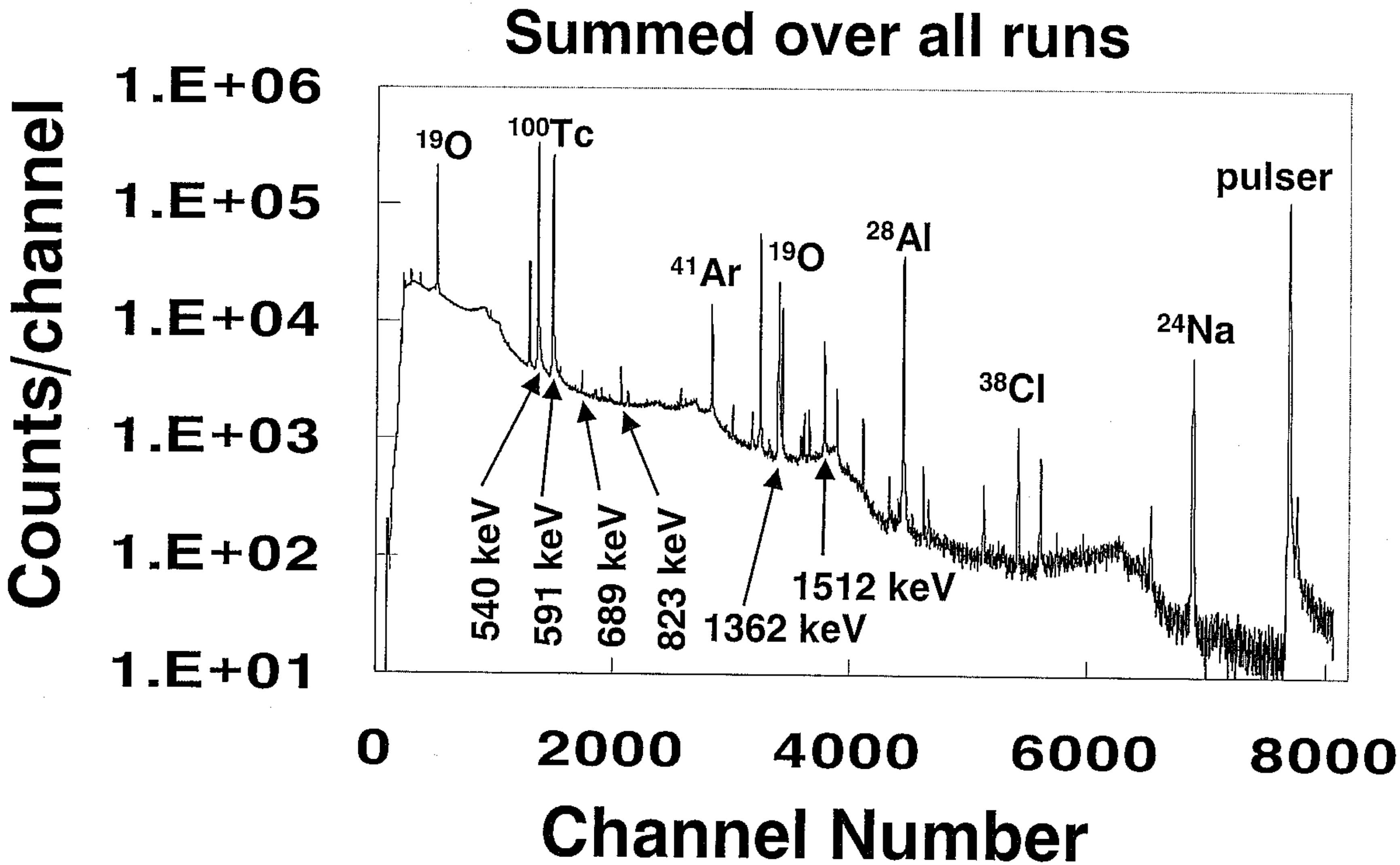
- in the data analysis,
  - β feedings to the g.s. and 5 major excited states are included in the analysis
  - the differences in ε_β for each β branch are taken into account

The β–γ coincidence measurement

- using a HPGe and a plastic β-ray detector (E_γ, E_β, TAC)
- *n* irradiation : KUR Pn-3 pneumatic tube
- Target : $NH_4{}^{99}TcO_4$ in NH_4OH (4.9 Bq) dried on Acrylic plates
- Irradiations : 15 s, 99 times



γ-ray spectrum



Analysis

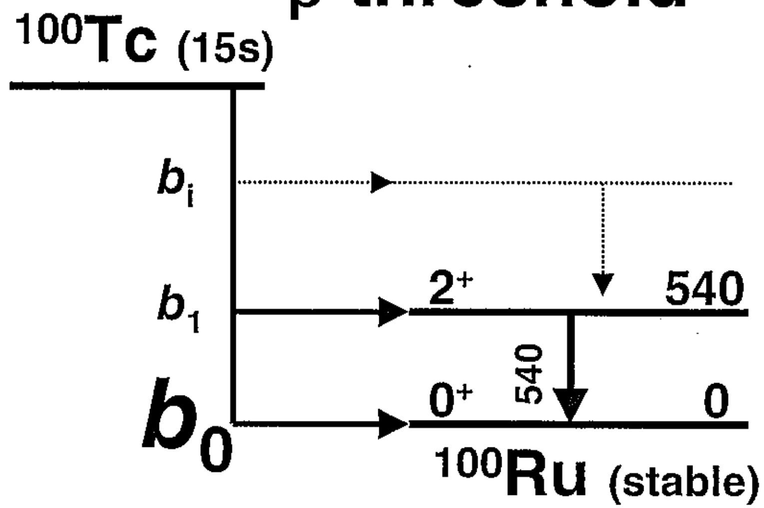
JNC TN8400 2000-028

Experimental result ⟷ **Comparison** ⟷ **Simulation using EGS4**

$$\frac{n_c(540\text{keV})}{n_\beta}$$

- TAC gates
- γ gates

- b_0 is varied
- β threshold



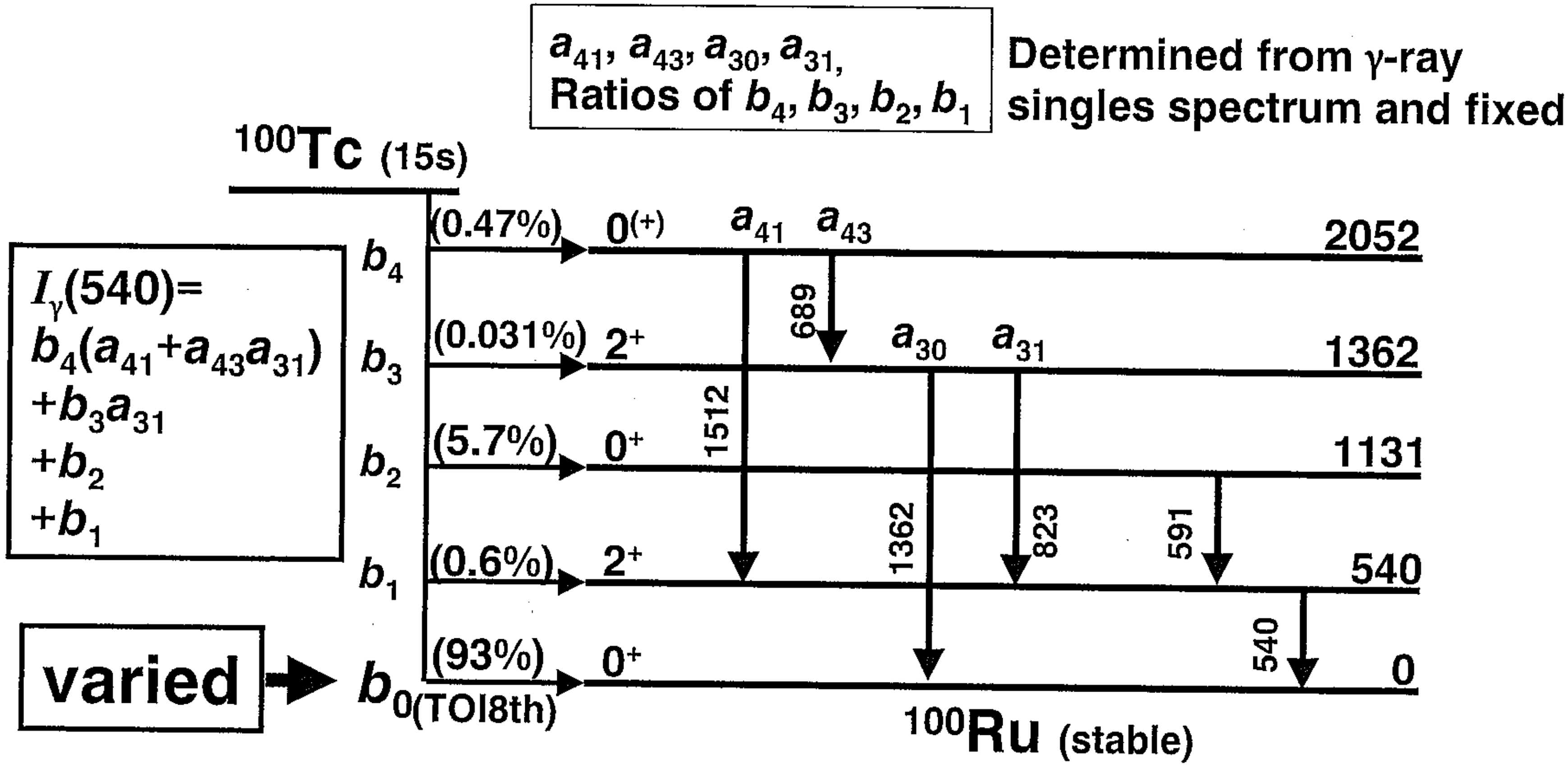
$[n_c(540\text{keV})/n_\beta]_{exp}$ =
0.00168 ± 0.00001
(Weighted average of all 99 runs)

n_c(540keV) : counting rate of β rays coin. With 540 keV γ rays
n_β : singles β-ray counting rate

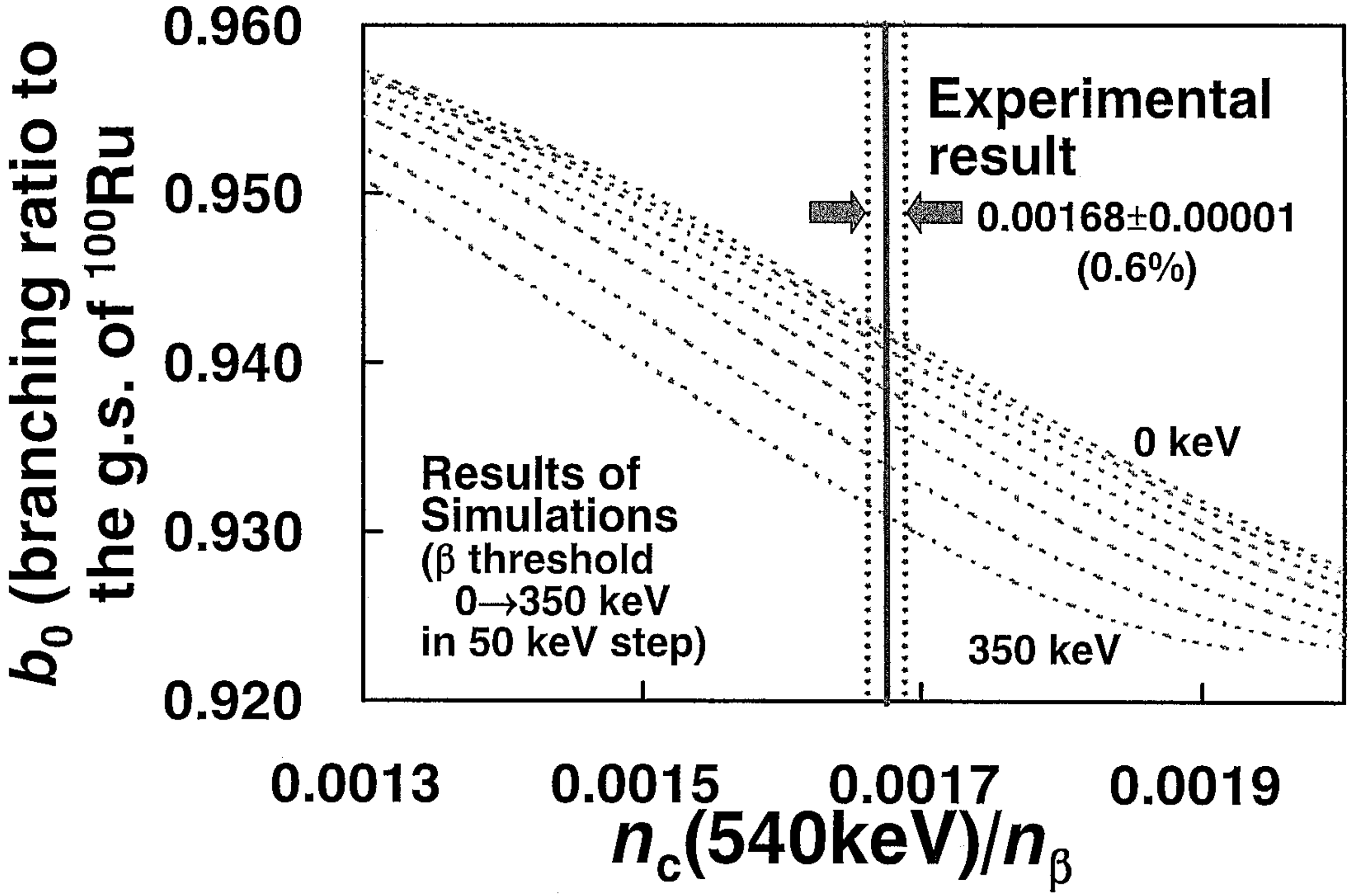
EGS4 Simulation of n_c(540keV)/n_β

▲ in which only the following levels/gammas are considered :

remaining β feedings < 0.24% (T.O.I. 8th ed)



Deduction of the branching ratio to the ground state



Summary

- To determine I_γ of ^{100}Tc accurately, a β−γ coincidence experiment has performed, using a HPGe and a plastic scintillator.
- The n_c(540keV)/n_β value was acculately determined to be 0.00168±0.00001.
- Analysis has been done in which β feedings to the excited levels and β detection threshold are taken into account.

Tasks remained

- Accurate determination of β detection threshold and the final results.
- Application of the method to other nuclides.

謝辞

本研究を行っていく上で、多くの方々にお世話になりました。

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