DISTRIBUTION OF PHOTON STRENGTH IN NUCLEI BY A METHOD OF TWO-STEP CASCADES DE91 005436

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ABSTRACT

The applicability of sum-coincidence measurements of two-step cascade γ -ray spectra to the determination of photon strength functions at intermediate γ -ray energies (3 or 4 MeV) is discussed. An experiment based on thermal neutron capture in Nd was undertaken at the Brookhaven National Laboratory High Flux Beam Reactor (BNL HFBR) to test this model. To understand the role of various uncertainties in similar experiments a series of model calculations was performed. We present an analysis of our experimental data which demonstrates the high sensitivity of the method to E1 and M1 photon strength functions. Our experimental data are in sharp contradiction to those expected from an E1 photon strength distributed according to the classical Lorentzian form with an energy invariant damping width. An alternative distribution of Kadenskij et al., which violates Brink's Hypothesis, is strongly preferred.

INTRODUCTION

In the past three decades a large amount of information has been accumulated from the decay of highly excited nuclear states, especially those states populated in slow neutron capture. This information concerns mainly hard primary γ rays that correspond to the transitions from the capturing states to low-lying levels. On the other hand, very little has been learned on soft primary γ rays with energies $E_\gamma \leq 4$ MeV, although such information is crucial for a better understanding of the deexcitation mechanism.

The radiative strength is usually treated in terms of the conventionally defined photon strength functions $S_\gamma(E_\gamma)$ for various multipolarities of γ radiation¹. The richest information is available for the E1 photon strength function. In order to calculate the sizes of total radiative widths of neutron resonances, Brink² used the principle of detailed balance and the concept of an electric Giant Dipole Resonance (GDR) built on the ground and each excited state. His values were typically a factor of three larger than those observed experimentally. Later, Axel³ used these assumptions to explain the energy variation of the $S_\gamma^{E1}(E_\gamma)$.

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McCullagh, Stelts, and Chrien⁴, in their review of E1 strength functions, pointed out that an energy-dependent damping width in the Lorentzian could improve the representation of the strength functions for low-energy γ rays. Subsequently, Kadenskij, Markushev, and Furman⁵ pointed out that for the cases of spherical and transitional nuclei the limit for $S_\gamma^{E1}(E_\gamma)$ as $E_\gamma \rightarrow 0$ should be non-zero. Using the Fermi liquid model these authors also predicted a specific dependence of width Γ_G of the electric GDR on the γ -ray energy, as well as a dependence of the shape of the GDR on the nuclear temperature of the final state on which the GDR is built. These considerations lead to a drastic modification of Brink's hypothesis and rule out a purely Lorentzian shape of the electric GDR.

In later papers, Kopecky and Chrien⁶ and Kopecky and Uhl⁷ used photoneutron data and slow and fast neutron capture data in ^{94}Nb , ^{106}Pd , ^{144}Nd , and ^{198}Au to support the above-outlined predictions of Kadenskij et al.⁵ about the shape and size of $S_\gamma^{E1}(E_\gamma)$. On the other hand, conclusions in Ref. 7, concerning the existence of the non-zero limit of $S_\gamma(E_\gamma)$, need verification that would lean on more direct information on photon strength at low energies. We note in this connection that an attempt to understand behavior of E1 and M1 photon strengths for low-energy primary transitions was made by Aldea and Seyfarth⁸ and Furman et al.⁹, who studied γ - α cascades in the $^{143}\text{Nd}(n,\gamma\alpha)^{140}\text{Ce}$ reaction.

In this paper we demonstrate that the method of Two-Step Cascade (TSC) γ - γ transitions, devised by Hoogenboom¹⁰ more than 30 years ago, makes the needed information available. We present results based on an experimental study of the $^{143}\text{Nd}(n,\gamma)^{144}\text{Nd}$ reaction and compare them with analogous results following from model calculations. We discuss the sensitivity of the proposed method to various models used for photon strength functions, and present conclusions concerning the best choice from these models for the reaction studied.

THE EXPERIMENT

The experiment is based on a two-detector coincident measurement of two-step cascades that follow thermal neutron capture and end at a final state of known spin and parity. By fixing an energy sum a final state can be selected and energy spectrum of all the transitions involved in TSC deexcitation can be obtained. This spectrum, called hereafter a TSC γ -ray spectrum, may be divided into two components: a set of discrete, well-resolved lines and an unresolved quasi-continuum. The discrete lines correspond to cascades proceeding via intermediate states with excitation energies below ≈ 3 MeV. The shape and size of the TSC spectrum carry information about the photon strength functions that govern the emission of γ rays in the deexcitation process. As the TSC spectra are expected to display a gross structure with a maximum at approximately 3 or 4 MeV, the behavior of the photon strength functions at these energies can be studied. A set of the TSC spectra can be obtained, one for each final state.

We have undertaken the above-described measurement for the thermal $^{143}\text{Nd}(n,\gamma)^{144}\text{Nd}$ reaction. A sample of natural Nd was irradiated in a beam of thermal neutrons at the BNL HFBR for about 300 hours. Two back-to-back HPGe detectors were used, having measured overall photopeak efficiencies of 1.95 and 2.30% at energy of 696 keV, including the effect of solid angle. Each event, represented by the deposited energies of the coincident γ rays and the associated detection time difference, was recorded on a magnetic tape. This information was later scanned off-line to yield the needed γ -ray spectra.

In the special scanning procedure used, a region that surrounds the peak corresponding to a preselected final state at which two-step cascades terminate is defined in the energy sum-time plane. The region is used to provide a background correction for a given TSC spectrum. This guarantees that the resulting TSC spectra represent a real effect, free from background, and that these spectra behave as if they were yielded by a spectrometer whose energy-response function is confined to a narrow single peak. The energy resolution achieved is represented by FWHM of 2.8 and 4.9 keV at energies of 600 and 7100 keV, respectively, including long-term instabilities.

The spectra obtained were corrected for energy variation of detector efficiencies and converted into spectra of *absolute* TSC intensities. A separate thermal (n,γ) single-spectrum measurement with a mixed Nd+Cl target has been undertaken to determine an absolute intensity for a primary transition to the $J^\pi = 4^+$ state at 1314 keV in ^{144}Nd . The result, $I_\gamma = 7.36\%$, has been used as a reference value in the normalization.

An example of a TSC spectrum is given in Fig. 1. It corresponds to all cascades terminating at the first $J^\pi = 2^+$ level in ^{144}Nd at 696 keV. The spectrum displays a symmetry that follows from the impossibility to distinguish between the primary and the secondary γ rays. A large number of discrete lines can be seen in Fig. 1. For some of them, energies of the corresponding intermediate ^{144}Nd states are given. Intensity fluctuations of these lines are very broad: the TSC intensity of the strongest line in the spectrum, corresponding to the $J^\pi = 4^+$ intermediate state at 1314 keV, reaches a peak value of 0.012 keV^{-1} , while a large number of still well-separated lines are weaker by a factor of 100.

The most important constituent of the spectrum is the quasi-continuum that shows up as a smear in a vicinity of the base line. In order to minimize the uncertainty due to the obvious fluctuations, an integral over a restricted energy interval, situated in the middle of the spectrum, can be chosen as a measure of the size of the TSC spectrum. We selected an energy interval of 2.4 MeV. In this case, the integral could be determined with a statistical accuracy of 2.6%. The integral of the TSC spectrum thus represents a positive, statistically-significant effect.

The integrals of TSC spectra were additionally corrected for effects of pair production and positron annihilation, as well as for backscattering of γ rays. These corrections are small: the overall change is about 6%. We also made corrections for effects of vetoing, caused by detection of those γ rays that were emitted after two-step cascades reached the fixed final state.

Fig. 1. The TSC γ spectrum for the $J^\pi = 2^+$ final state of ^{144}Nd at 696 keV. Only a small fraction of the range of the TSC intensity is shown.

Vetoing correction factors range from 0.73 to 1.39. No corrections were done for bremsstrahlung. Its contribution to the useful part of a TSC spectrum is expected to be small, not higher than 2%.

All TSC spectra obtained are subject to a common systematic error of 16%, associated with uncertainties of the above-outlined corrections and normalization.

In a similar manner, TSC γ -ray spectra were obtained also for the $J^\pi = 0^+$ ground state and for the states at 1314, 1510, and 1561 keV with J^π equal to 4^+ , 3^- , and 2^+ , respectively. The integrals of these spectra could be determined with a statistical accuracy of 9.7, 6.8, 7.2, and 12.4%, respectively.

MODELING

In order to test various hypotheses about photon strength functions we modeled the TSC process and compared the resulting spectra with those observed experimentally.

Let us assume a cascade $EJ\pi \rightarrow E'J'\pi' \rightarrow E''J''\pi''$. Here, $EJ\pi$ stands for an excitation energy, spin and parity of the initial state. Similarly, $E'J'\pi'$

and $E''J''\pi''$ belong to the intermediate state and the fixed final state, respectively. Following the definition of the photon strength function the expectation value of a partial radiative width for the first step $EJ\pi \rightarrow E'J'\pi'$ can be expressed as¹

$$\langle \Gamma_{\gamma}(EJ\pi \rightarrow E'J'\pi') \rangle = E_{\gamma 1}^{2L+1} S_{\gamma}^{XL}(E_{\gamma 1}) / \rho(E, J), \quad (1)$$

where XL stands for multipolarity of the transition $EJ\pi \rightarrow E'J'\pi'$, i.e. $\lambda L = E1, M1, E2$, etc., $E_{\gamma 1}$ is a γ -ray energy of this transition and $\rho(E, J)$ - density of levels with spin J at an excitation energy E .

The expectation value of the total radiative width for the state $EJ\pi$ is

$$\begin{aligned} \langle \Gamma_{\gamma EJ\pi} \rangle = & \sum_{J'\pi'} \int_{E_0}^E \rho(E', J') \langle \Gamma_{\gamma}(EJ\pi \rightarrow E'J'\pi') \rangle dE' \\ & + \sum_k \langle \Gamma_{\gamma}(EJ\pi \rightarrow E_k J_k \pi_k) \rangle. \end{aligned} \quad (2)$$

The first term of this expression includes the continuum of states with $E' > E_0$ that are described by level density ρ . The second term belongs to transitions to the remaining discrete states, labeled by subscript k . It is assumed that below the energy E_0 a full set of states is known from experiments. Analogous expressions can be written for $\langle \Gamma_{\gamma}(E'J'\pi' \rightarrow E''J''\pi'') \rangle$ and $\langle \Gamma_{\gamma E'J'\pi'} \rangle$.

The expectation value of a TSC intensity, per unit energy interval, can be expressed via expectation values of one-step intensities:

$$\langle I_{\gamma\gamma}(E_{\gamma 1}) \rangle = \sum_{J'\pi'} \langle I_{\gamma}(EJ\pi \rightarrow E'J'\pi') \rangle \langle I_{\gamma}(E'J'\pi' \rightarrow E''J''\pi'') \rangle \rho(E', J'). \quad (3)$$

Here $E' = E - E_{\gamma 1}$ and

$$\langle I_{\gamma}(EJ\pi \rightarrow E'J'\pi') \rangle \simeq \langle \Gamma_{\gamma}(EJ\pi \rightarrow E'J'\pi') \rangle / \langle \Gamma_{\gamma EJ\pi} \rangle. \quad (4)$$

A similar expression can be written for $\langle I_{\gamma}(E'J'\pi' \rightarrow E''J''\pi'') \rangle$. Equation 3 is valid for primary γ -ray energies $E_{\gamma 1} < E - E_0$. The introduced quantity $\langle I_{\gamma\gamma}(E_{\gamma 1}) \rangle$ is an energy density, i.e. it represents an intensity per unit of energy.

Examples of calculated TSC spectra are given in Fig. 2. Besides the values of $\langle I_{\gamma\gamma}(E_{\gamma 1}) \rangle$ the values of individual terms of the right-hand side of Eq. 3 for various $J'\pi'$ are plotted. The spectra in Fig. 2 have been calculated using a level density formula of Kataria et al.¹¹ with parameters, recommended by Kopecky and Uhl⁷. The photon strength function S_{γ}^{E1} used in calculations was deduced from the model of Kadenskij et al.⁵

Fig. 2. Expectation values of the TSC intensity vs γ -ray energy for the $J^\pi = 2^+$ state of ^{144}Nd at 696 keV (solid line). Contributions of intermediate states with individual values of J^π are also shown.

For $S_\gamma M1$ and $S_\gamma E2$ single-particle approximations were assumed. The role of $M2$, $E3$, and higher multiplicities was neglected.

In order to study the influence of Porter-Thomas fluctuations of partial radiative widths we modified Eq. 3 to yield fluctuating values of TSC intensity. Specifically, in Eq. 3 we introduce additional factors $\theta_{E'J'\pi'}^{(1)}$, and $\theta_{E'J'\pi'}^{(2)}$, that are responsible for Porter-Thomas fluctuations of one-step intensities. The fluctuating TSC intensity can be expressed as

$$I_{YY}^{(\text{fluct})}(E_{Y1}) = \sum_{E'J'\pi'} \langle I_Y(EJ\pi \rightarrow E'J'\pi') \rangle \langle I_Y(E'J'\pi' \rightarrow E''J''\pi'') \rangle \times \theta_{E'J'\pi'}^{(1)} \theta_{E'J'\pi'}^{(2)} \delta(E_{Y1} - E + E'). \quad (5)$$

Summation in Eq. 5 is assumed over a full set of all the intermediate states, whose values of excitation energy E' are distributed in accordance with the adopted level-density formula.

Fig. 3. Calculated and measured TSC intensities for the individual states of ^{144}Nd . The calculated values were obtained using the choice (a) for $S_\gamma^{E1}(E_\gamma)$, see text. The upper part: a magnetic GDR, $\langle\Gamma_\gamma\rangle = 157$ MeV. The lower part: a constant strength $S_\gamma^{M1} = 1.2 \times 10^{-8} \text{ MeV}^{-3}$, $\langle\Gamma_\gamma\rangle = 195$ MeV.

It can be shown that products $\theta_{E'\pi}^{(1)}, \theta_{E'\pi}^{(2)}$, behave as completely independent statistical variables, obeying a distribution which can be easily reproduced by the method of Monte Carlo. This makes it possible to model the values of $I_\gamma^{(\text{fluct})}$.

In order to estimate the role of Porter-Thomas fluctuations we used Eq. 5 and modeled TSC spectra for various final states. All TSC intensities $I_\gamma^{(\text{fluct})}$ we obtained exhibit violent fluctuations, even when they are represented as histograms with a bin width of 3 keV to resemble spectra obtained by a typical Ge-detector. Nevertheless, if one integrates the values of $I_\gamma^{(\text{fluct})}$ over a reasonably wide energy interval, fluctuations of the resulting integral can be kept low. Specifically, for the case of the earlier-mentioned energy interval of 2.4 MeV the residual fluctuations of integrated TSC intensities are represented by rms values of 23, 11, 10, 9, and 11% for cascades populating the ground state and the states at 696, 1314, 1510, and 1561 keV, respectively. These rms values were obtained using the same level-density formula and the same explicit expressions for

Fig. 4. Calculated and measured TSC intensities for the individual states of ^{144}Nd . The calculated values were obtained using the choice (b) for $S_\gamma^{E1}(E_\gamma)$, see text. The upper part: a magnetic GDR, $\langle \Gamma_\gamma \rangle = 17$ MeV. The lower part: a constant strength $S_\gamma^{M1} = 0.3 \times 10^{-8}$ MeV $^{-3}$, $\langle \Gamma_\gamma \rangle = 25$ MeV.

photon strength functions as in the case mentioned in connection with Fig. 2. We see that the quoted experimental uncertainties in the determination of the quasi-continuum are comparable to, or even lower than those following from the Porter-Thomas fluctuations.

Quantities $\langle I_{\gamma\gamma} \rangle$ and $I_{\gamma\gamma}(\text{fluct})$ were modeled for the ^{144}Nd ground state and the levels at 696, 1314, 1510, and 1561 keV. Using Eq. 2 we also calculated expectation values of total radiative widths for an excitation energy E adjusted at the neutron threshold and for the value $J_\pi = 3^-$ that belongs to the dominating capturing state in the reaction studied. The results are presented in a condensed form in Figs. 3-5, where the integrated TSC intensities are plotted for individual levels and compared with analogous quantities deduced from the measured TSC spectra. An energy interval of 2.4 MeV was selected for integration. In Figs. 3-5 experimental errors of the TSC intensities are plotted as well as uncertainties associated with Porter-Thomas fluctuations.

Fig. 5. Calculated and measured TSC intensities for the individual states of ^{144}Nd . The calculated values were obtained using the choice (c) for $S_\gamma^{E1}(E_\gamma)$, see text. The upper part: a magnetic GDR, $\langle \Gamma_\gamma \rangle = 53 \text{ MeV}$. The lower part: a constant strength $S_\gamma^{M1} = 0.6 \times 10^{-8} \text{ MeV}^{-3}$, $\langle \Gamma_\gamma \rangle = 72 \text{ MeV}$.

The presented results are based on the use of the level-density formula of Kataria et al.¹¹ and the following three approximations for the E1 photon strength function:

- The expression based on a classical electric GDR of Lorentzian shape.
- The expression (a) modified by inclusion of an energy- and temperature-dependent damping width⁶, specifically

$$\Gamma_G = \Gamma_G^{(0)} (E_\gamma^2 + 4\pi^2 T^2) / E_G^2, \quad (6)$$

where T is the nuclear temperature of the level on which an electric GDR is built, $\Gamma^{(0)}$ --a constant factor and E_G --the GDR energy.

- The expression, representing a low-energy approximation within the model of Kadenskij et al.⁵, i.e.

$$S_{\gamma}^{E1}(E_{\gamma}) = \frac{(1+2f_1'/3)^{1/2} \sigma_0 \Gamma_G^{(0)} \Gamma_G E_G}{3\pi^2 \hbar^2 c^2 (1+2f')^{1/2} (E_{\gamma}^2 - E_G^2)^2}, \quad (7)$$

where σ_0 is the photoabsorption cross section at peak of the GDR, Γ_G is given by Eq. 6 and f_1, f' are parameters of the Fermi-liquid model that characterize a quasi-particle interaction. In accordance with Ref. 12 we took $f_1' = -0.1$ and $f' = 0.25$.

We made several assumptions on the M1 strengths; first, a single-particle model assuming various strengths and, second, a magnetic giant dipole resonance model with parameters taken from Ref. 7. The best fits are shown in Figs. 3-5.

DISCUSSION AND SUMMARY

As can be seen from Fig. 3, the choice (a) is not adequate. Much better agreement between the data and model calculations is observed in Figs. 4 and 5 for the remaining two choices of S_{γ}^{E1} . A good agreement is achieved for the photon strength function that is represented by the approximation of Kadenskij et al.⁵, while S_{γ}^{M1} is assumed to be energy-independent, see Fig. 5. This accordance between the measured and calculated TSC intensities can be further improved by a slight adjustment of the measured intensities within their quoted common uncertainty of 16%. After such adjustment the agreement between the data and calculations can be characterized by a χ^2 -value of 5.2 at $\nu = 4$. It should be stressed that the calculated expectation value of the partial radiative width $\langle \Gamma_{\gamma} \rangle = 72$ MeV is reasonably close to the experimental value $\Gamma_{\gamma}^{(exp)} = 80 \pm 9$ MeV (see Ref. 13). If we assume that S_{γ}^{M1} is energy-independent the choice (b) also gives a good agreement between the calculated and measured TSC intensities, see Fig. 4. However, in this case the model calculations yield a value $\langle \Gamma_{\gamma} \rangle = 25$ MeV which is too low to accept the considered choice for E1 photon strength function as correct.

Compared to other methods, the most important advantage of the method used is its inherent sensitivity to the photon strength of primary transitions as well as to that of secondary transitions. For the choice (c) that violates Brink's hypothesis, these strengths are substantially different both in size and their energy dependence, while in case of validity of this hypothesis the strengths are identical. For this reason the conclusion we made about the validity of the choice (c) represents a meaningful verification of the predicted violation of Brink's hypothesis.

A closer inspection of Figs. 3-5 reveals that a crucial role in testing various models is played by the integrated TSC intensities for the cascades ending at the negative-parity 1510 keV state.

Under certain simplifying assumptions one can find from Eqs. 1-3 that for $S_{\gamma}^{E1} \approx S_{\gamma}^{M1}$ at energies $E_{\gamma} \approx 3$ or 4 MeV the integrated TSC intensities will not depend on the parity π of the final state. However, for S_{γ}^{E1} significantly different from S_{γ}^{M1} the TSC intensities for the cascades

ending at states with $\pi'' = \pi$ will be enhanced. By this important feature the method of two-step cascades differs substantially from previous methods that are sensitive mostly to the sum of S_Y^{E1} and S_Y^{M1} .

In the majority of cases the deviations between the modeled and measured integrated TSC intensities are much higher than experimental errors and uncertainties associated with Porter-Thomas fluctuations. The observed deviations are statistically meaningful. This demonstrates that the TSC intensities are highly sensitive to the choices of various explicit expressions for photon strength functions.

In conclusion, it is evident that the method of two-step cascades represents a promising tool. Experiments similar to that described in this paper could be repeated for a large number of nuclei with a perspective of a deeper understanding of photon strength distributions.

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