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Validating resonance properties using nuclear resonance fluorescence

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Measurement of a resonance's integrated cross section using nuclear resonance fluorescence can be a valuable tool for verifying the properties of the resonance because of the clear and unambiguous physical connection to the spin, lifetime, and ground state branching ratio of the level. We demonstrate this idea by measuring the integrated cross section of the 3.004-MeV level in ^{27}Al to 4% using the monoenergetic γ-ray beam at the High Intensity γ-ray Source. That level was the subject of much debate experimentally in the 1960s, especially its spin, and even now only has a current tentative spin assignment of $J = (9/2)$. The consistency check between this integrated cross section and the known properties of the level indicate that one (or more) of the literature properties is incorrect. Based on the range of extent of each property, a reassignment of spin to a tentative $J = (7/2)$ may be warranted, but this would need to be confirmed with other measurements. This result demonstrates the utility of NRF as a way to verify the properties of states in the literature before undertaking more extensive measurements.

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I. INTRODUCTION

Compiled nuclear structure data is used in a wide variety of applications including reaction rate calculations, nuclear astrophysics, nuclear reactor simulations, and homeland security. It also plays a prominent role in developing systematics and verifying theoretical nuclear models. The particularly important and relevant structural data for individual levels are the spin, parity, lifetime, and ground state branching ratios. In many cases, however, there are conflicting results and incomplete information for a given level. A method which could, even for limited cases, provide a way to readily validate resonance data could be helpful to establish the confidence of nuclear data used for a particular application, and help ascertain which levels are in need of additional measurements.

Nuclear resonance fluorescence (NRF) is one such method that could validate resonance properties. It is a process whereby the nucleus is resonantly excited by absorption of a γ ray and decays by emitting a γ ray either back to the ground state or to an excited state. The NRF integrated cross section is sensitive to the level spin because it is directly proportional to the statistical spin factor g. It is also sensitive to the level's lifetime and ground state branching ratio. For excitation from, and decay to, the ground state, the integrated cross section is [1]

$$I_{\rm cs} = g\left(\frac{\pi\hbar c}{E_\gamma}\right)^2 b_0^2\Gamma, \qquad (1)$$

where $g = (2J_f + 1)/(2J_i + 1)$, and J_f is the spin of the excited level, and J_i is the spin of the ground state, E_γ is the energy of the level, Γ is the level width (inversely proportional to the lifetime), and b_0 is the ground state branching ratio. Changes in the level spin by a single unit of angular momentum results in a significant change in the predicted integrated cross section providing a unique sensitivity to the spin. The integrated cross section is not directly affected by the parity of the level. NRF provides a strong consistency check between the several properties. The physics of resonance excitation is well established suggesting its use as a validation method for nuclear data.

NRF is particularly useful because of the relative simplicity of measuring and analyzing the data. Target preparation is simplified as measurements can be done at atmosphere, and the target material can be in any form including liquid or gas. Adequate data can be acquired in as little as one hour, or less, depending on the experiment design, and the data analysis can be completed rapidly for certain situations, as discussed in the experimental section below. It is constrained, however, to being useful for levels with a large ground state branching ratio, and a reasonably short lifetime.

The use of NRF as a validation tool is demonstrated in the present work by attempting to validate the resonance data for the 3.004-MeV level in ^{27}Al. The spin assignment of the 3.004-MeV level has a significant impact on the interpretation of the low-energy structure of ^{27}Al, but is currently only given a tentative assignment. Experiments reported spin values of $J = 9/2$ [2,3] (neutron scattering and γ-ray angular correlation, respectively), $J = 7/2$ [4] (triple γ-ray angular correlation), and $J = 5/2, 9/2$ [5,6] (γ-ray angular correlation). Preference for $J^\pi = (9/2^+)$ is given in the nuclear data evaluation [7], as well as in the literature [8] (positive parity is supported by the experimental results—as such, parity will be dropped for the remainder of the paper). In comparison, the lifetime (85 ± 4 fs; see Table 27.8 in Endt [9] for a summary of measurements) and ground state branching ratio (88^{+2}_{-5}%) of this level are well known [7].

Theoretically, a spin assignment of $J = 9/2$ is supported as the nuclear shell model calculations in the sd-shell region using the "universal" s-d interaction predict a spin $J = 9/2$ level within 100 keV of the 3.004-MeV level [10]. The original (and older) strong coupling (Nilsson) model, in

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contrast, predicted a level with $J = 7/2$ near 3.0 MeV [11,12]. The proposed $J = 9/2$ assignment for the 3.004-MeV level was the most problematic issue for this model. Most other experimental phenomena, however, were adequately explained by the strong coupling model [8,13].

By accurately measuring the level's NRF integrated cross section the proposed resonance properties can be validated. The present data validation is possible because of the present order-of-magnitude improvement in the uncertainty of the integrated cross section of the 3.004-MeV level over previous NRF measurements of ^{27}Al.

^{27}Al presents a unique case where the properties of levels near the 3.004-MeV level (particularly the 2.982-MeV level) are well known, but that of the 3.004-MeV level is less-firmly established because of its longer lifetime and its close proximity to the 2.982-MeV level. ^{27}Al has the additional benefit for demonstrating the current method in that the NRF integrated cross sections have been measured in two previous NRF experiments: that of Vodhanel *et al.* [14] and of Pietralla *et al.* [15], the latter one having established the lifetime of the nearby states to high precision. Both experiments, however, used bremsstrahlung beams which produce a large background limiting the ability to accurately measure the integrated cross section of states with a relatively long lifetime such as that of the 3.004-MeV level in ^{27}Al. Those papers could only limit the integrated cross section to 5.6 ± 3.3 eV b, and 3.9 ± 1.7 eV b (assuming $J = 9/2$), respectively. The high-precision results for the larger resonances in ^{27}Al by Pietralla *et al.* have been widely used as the standard for flux calibration in NRF experiments, and provide a solid reference for determining the integrated cross section of the 3.004-MeV level relative to. Specifically, the 2.982-MeV level's integrated cross section was determined at 2.1%, being 31.9 ± 0.7 eV b.

To enable a high-precision measurement of the integrated cross section for the 3.004-MeV level, we used a quasimonoenergetic γ-ray beam to eliminate the beam-induced background (see, e.g., Ref. [16]). This reduced the uncertainty of the final integrated cross section to 4%. We discuss how only if a spin of $J = 7/2$ is considered are the 3.004-MeV level's more firmly established lifetime and ground state branching ratio consistent with the presently measured integrated cross section. We additionally discuss the counter cases for the possibilities for the lifetime or branching ratio being incorrect.

II. EXPERIMENT

The ^{27}Al 3.004-MeV level's integrated cross section was precisely determined from a ratio to that of the well-known, and simultaneously excited, 2.982-MeV level which reduced this experiment's systematic uncertainties. The levels were simultaneously excited by NRF using the high-flux ($\approx 10^8$ γ/s) circularly polarized quasimonoenergetic γ-ray beam at the High Intensity γ-ray Source (HIγS) at Triangle Universities Nuclear Laboratory in Durham, NC, USA [16]. The present NRF measurement of a light target is similar in conditions to those used to measure NRF of levels in ^{11}B [17].

The NRF signature was measured by placing a 2.53-cm-thick Al scattering target in the middle of four 60% HPGe detectors which were an average of 12 cm from the target, and

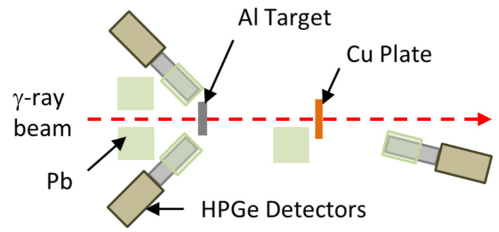

FIG. 1. (Color online) The experimental setup (distances are not to scale). The NRF scattering signal was observed using four detectors. The two detectors out of the plane of the image are not shown. The monitor detector observed Compton scattering off of a Cu plate which was used to correct for variations in the beam profile during measurement.

at a scattering angle of $\theta = 120^\circ$, with two in the horizontal plane, and two in the vertical plane (see Fig. 1). They were shielded with 2.5-cm Pb on the sides, and Pb (0.55 cm) and Cu (0.4 cm) filters were placed in front of the detectors to reduce the intensity of low energy photons hitting the detector. The detector efficiencies were measured using a calibrated 9.16 μCi ^{56}Co source.

A HPGe monitor detector placed further downstream of the beam was used to measure the beam profile directly when placed in the beam periodically during the experiment (the flux was reduced during these measurements by lowering Cu attenuators into the beam further upstream). It was also used to monitor for slight changes in the beam profile during active NRF measurement by measuring the Compton scattering off of a Cu plate positioned after the Al target (see Sec. II A for full details). During part of the experiment there was Pb shielding placed next to the Cu plate to prevent Compton scattering from the Al target from also reaching the HPGe detector, and during the rest of experiment the Pb shielding was absent, allowing the Compton scattering peaks from both the Al target and the Cu plate to reach the detector.

Since the levels were simultaneously excited (see Fig. 2), the integrated cross section of the 3.004-MeV level (superscript B) can be determined as a ratio to the integrated cross

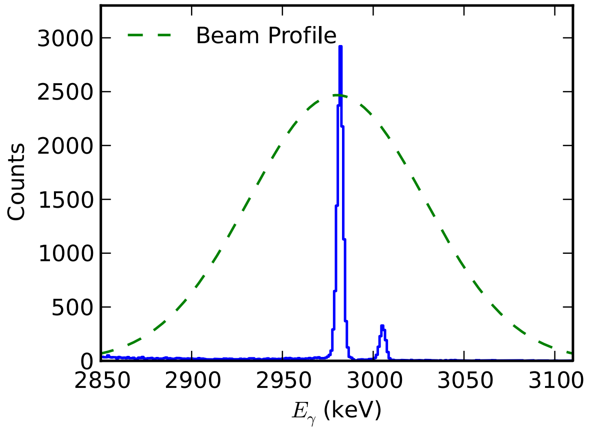

FIG. 2. (Color online) The γ-ray spectrum of the de-excitation of the 2.982- and 3.004-MeV levels of ^{27}Al observed using NRF with the beam profile overlaid.

section of the 2.982-MeV level (superscript A) [1]:

$$I_{cs}^{B} = I_{cs}^{A} \frac{N^{B}}{N^{A}} \frac{\phi^{A}\epsilon^{A}\lambda_{tt}^{A}}{\phi^{B}\epsilon^{B}\lambda_{tt}^{B}}. \quad (2)$$

The peak areas (N)—determined by fitting with a Gaussian—are corrected for the relative amount of flux (or beam profile, ϕ), the efficiency (ϵ), and for the thick target correction (λ_{tt}). All other factors, including pileup, detector livetime, and absolute flux, cancel out in the ratio. This ratio method has been used previously to determine the integrated cross section of resonances [18]. Though the difference in the efficiency at the two energies is corrected for, the difference is only about 0.6%. Similarly, the difference in the beam profile between the two energies ranged from about 0.1% to just less than 4%. The thick target correction is significant, but can be calculated analytically and accurately using the known resonance parameters from Pietralla *et al.* [15], as discussed below.

The ratio method provides a straightforward and rapid way to measure an integrated cross section and validate nuclear data. If a well-known, nearby resonance is not available for a given nucleus, the NRF measurement can be readily done by including a second target behind the primary target of a material which does contain a relevant resonance. If the resonances are close in energy, and the targets are thin, then the several corrections mentioned above can be neglected, and the ratio of the peak areas multiplied by the known integrated cross section determines the unknown integrated cross section.

NRF can also be used to directly measure the spin of a state from the angular distribution of emitted γ rays when a linearly polarized beam is used, but only when the ground state spin of the target nucleus is $J = 0$ [16]. This was not possible in the present measurement because the ^{27}Al ground state spin is $J = 5/2$. Furthermore, we used a circularly polarized beam to optimize the flux.

Several measurements were made, 11 in total, nine of which included absorbers placed in the beam upstream, and six of those measurements used absorbers containing Al. The Al absorber thicknesses were either 2.537 ± 0.001 cm (thick) or 0.25 ± 0.05 cm (thin) of Al. The purpose of the absorbers was for an unrelated application study, and do not influence the present results, except for those measurements where an absorber containing Al was used. In those cases, the resonance absorption due to Al in the absorbers was corrected for by using the known lifetimes. For the 3.004-MeV level, there is a 1% difference in the absorption amount for different possible spins for the level, having little impact on the present measurement. The two no-absorber measurements each lasted one hour and yielded high statistics (about 20 000 counts in the 2.982-MeV peak and 2000 counts in the 3.004-MeV peak) demonstrating the relatively rapid measurement times that can be achieved.

A. Beam profile measurement

To improve the precision of the measurement, the beam energy profile was measured with a HPGe detector placed directly in the beam, and fitting the primary peak with a Gaussian. The beam intensity was reduced for the duration of this measurement by inserting Cu absorbers into the beam further upstream. The nonfull energy peak portion of the detector response (namely the Compton edge) was accommodated by fitting it with a Gaussian, used for a similar situation in Ref. [19]. Approximating the Compton edge underlying the full energy beam peak as the tail of a Gaussian is supported by noting that the Compton edge has an approximately Gaussian shape. Both Gaussians were fit simultaneously over a region from about 2.6 to 3.2 MeV (see Fig. 3). This yielded a result equivalent to using a response function to unfold the spectrum (see, e.g., Fig. 3 in Ref. [20]).

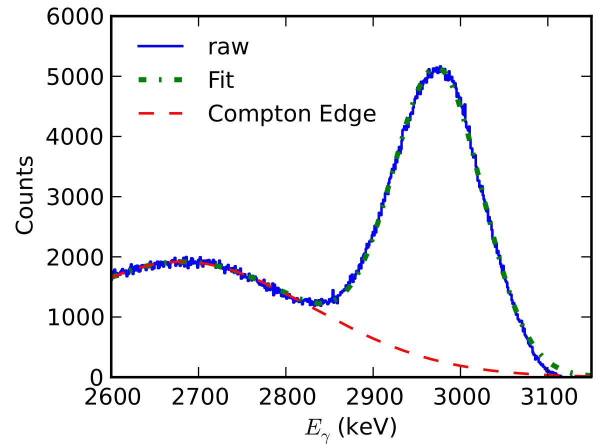

FIG. 3. (Color online) The measured γ-ray beam profile with fit overlaid.

The beam profile was also monitored online using the same detector by measuring the Compton scattering at about 9° off of a 1-mm-thick Cu plate. Small changes in the beam energy profile were compensated for by correlating the shape and position of the Compton-scattered peak with those of the directly measured beam profile peak. The parameters for the correlation were determined by measuring the Compton scattering peak, for approximately an hour, either immediately before or after the beam profile measurements in order to minimize the possibility of the beam parameters changing between the two measurements. To subtract the Compton scatter peak from the Al target, a reference measurement of that component was made with no Cu plate in place.

To determine the Compton scattered peak shape, the spectrum was deconvolved. This was done using a detector response matrix constructed by calibrating the semiempirical response model in Ref. [21] using a calibration spectrum measured by placing a ^{56}Co source on the Cu plate. Following deconvolving, the residual peak was fit with a Gaussian. Compensating for changes in the beam profile in this way reduced the associated systematic uncertainty from 3% to as low as 1.5%. In all cases, the pileup component of the spectrum was calculated and subtracted before fitting was done using the method described in Ref. [22].

B. Thick target correction

The thick target correction λ_{tt} incorporated both the beam attenuation due to nonresonant atomic processes as well as resonance absorption in the scattering target. It also included the effect of resonance absorption in the target for the cases in which an absorbing target containing Al was placed upstream. It was calculated analytically, accounting for resonance width and thermal broadening [1,23]:

$$\lambda\big(E,N_t^w;\Gamma_0\big) = \frac{1-\exp\big\{-[\sigma_D(E;\Gamma_0)+\sigma_e(1+1/\cos\theta)]N_t^w\big\}}{[\sigma_D(E;\Gamma_0)+\sigma_e(1+1/\cos\theta)]N_t^w}, \quad (3)$$

where σ_D is the thermally broadened resonance shape, Γ_0 is the resonance ground state width, σ_e is the atomic scattering cross section [24], N_t^w is the areal density of the scattering target, and θ is the scattering angle. Equation (3) is the thick target correction for the resonance shape and must be integrated over the energy E to obtain the scalar value λ_{tt}. This equation, for cases where there is an Al absorber upstream, is multiplied by $e^{-\sigma_D(E;\Gamma_0)n_a}$ before being integrated, where n_a is the Al absorption target areal density. The equation for σ_D can be found in Refs. [1,23].

For the cases where no Al was present upstream, $\lambda_{tt}^A = 0.62$ for the 2982-keV resonance, and $\lambda_{tt}^B = 0.74$ for the 3.004-MeV resonance, which included the contribution to λ_{tt} from atomic processes (σ_e term) in the scattering target. The contribution from atomic processes amount to 0.76, accounting for most of the thick target correction. For a thick (thin) Al absorber upstream, λ_{tt}^A was further multiplied by 0.66 (0.96). λ_{tt}^B was similarly multiplied by 0.95 (0.99).

C. Systematic uncertainties

For each integrated cross section, we determined the statistical and systematic uncertainties. The statistical uncertainty is determined from fitting the NRF peak. The ratio method's systematic uncertainty was split into two components: a reducible uncertainty, and an irreducible uncertainty.

The reducible uncertainty was that which randomly varied run-to-run, being the fluctuation in the beam profile ratio ($\frac{\Delta\phi}{\phi} = 1.5\%$), assessed from the plausible variation in the ratio with known variations in the peak position. For runs in which the Pb shielding next to the Cu plate was absent, the systematic uncertainty increased due to uncertainty that came from the necessity of subtracting the Compton scattering peak from the Al target in the monitor detector spectrum. This increased the systematic uncertainty to $\frac{\Delta\phi}{\phi} = 2.4\%$ on average for the affected runs.

The irreducible uncertainty was due to normalizing to the 2.982-MeV level's width ($\frac{\Delta I_{cs}}{I_{cs}} = 2.1\%$), and the uncertainty from the thick target correction ($\frac{\Delta\lambda}{\lambda} = 1.6\%$), which was also due to, predominantly, the uncertainty in the level width. Other contributions of uncertainty to the thick target correction due to imprecise knowledge in target or detector geometries cancel out because a ratio of the two resonances is taken. The uncertainty in the ratio of the two thick target corrections was studied for large changes in scattering angle ($\pm 10^\circ$) and was found to vary only negligibly (0.2%).

III. RESULTS

Using the present method, the integrated cross section of the 3.004-MeV level has been determined to 4% uncertainty (see Table I): $3.30 \pm 0.04 \pm 0.02 \pm 0.09$ [errors are statistical, systematic (reducible), and systematic (irreducible)].

TABLE I. The integrated cross section for the 3.004 MeV in ^{27}Al level determined from a ratio to the 2.982-MeV level for several runs. It was corrected for attenuation in an upstream Al absorber when applicable (runs marked with a † or ‡ for a thin or thick Al absorber, respectively). The uncertainties are given (in order) as statistical, systematic (reducible), and systematic (irreducible). The weighted mean of the experimental I_{cs} and the calculated I_{cs} for the several possible spin values are given at the bottom of the table.

Run	I_{cs} (eV b)
1	$3.36 \pm 0.13 \pm 0.05 \pm 0.09$
2	$3.28 \pm 0.16 \pm 0.08 \pm 0.09$
3	$3.34 \pm 0.11 \pm 0.08 \pm 0.09$
4	$3.30 \pm 0.19 \pm 0.07 \pm 0.09$
5	$3.35 \pm 0.18 \pm 0.08 \pm 0.09$
6†	$3.31 \pm 0.18 \pm 0.10 \pm 0.09$
7†	$3.24 \pm 0.17 \pm 0.05 \pm 0.09$
8†	$3.18 \pm 0.17 \pm 0.05 \pm 0.09$
9†	$3.29 \pm 0.14 \pm 0.07 \pm 0.09$
10‡	$3.26 \pm 0.12 \pm 0.05 \pm 0.11$
11‡	$3.41 \pm 0.12 \pm 0.07 \pm 0.11$
Mean	$3.30 \pm 0.04 \pm 0.02 \pm 0.09$
$J = 5/2$	$2.6^{+0.2}_{-0.3}$
$J = 7/2$	$3.4^{+0.2}_{-0.4}$
$J = 9/2$	$4.3^{+0.3}_{-0.5}$

This integrated cross section can be used to validate the data for the 3.004-MeV level. Given that the spin is the least-known property of the 3.004-MeV level, the expected integrated cross was calculated using Eq. (1) and the three spin values covering the range of previously reported spins: $J = 5/2$, $7/2$, and $9/2$ (see Table I, last three rows). The statistical spin factors are $g = 1.00$, 1.33, and 1.67, respectively. The level lifetime (width) and ground state branching ratio were taken from Ref. [7]. The spin value preferred by evaluation and theory, $J = 9/2$, yields an integrated cross section of $I_{cs} = 4.3^{+0.3}_{-0.5}$ eV b. There is a larger lower limit on the uncertainty (see Table I) reflecting the ground state branching ratio if the upper limit for unobserved branches are included. Even with this uncertainty, this result clearly excludes the present measurement indicating that the resonance properties are not consistent. The present integrated cross section is consistent with that calculated if $J = 7/2$ is used which gives $I_{cs} = 3.4^{+0.3}_{-0.4}$ eV b.

The alternate possibility of either the 3.004-MeV level's lifetime or branching ratio (and not the spin) being incorrect should also be considered. The inferred lifetime from the present results, assuming the literature values for spin ($J = 9/2$) and ground state branching ratio ($88^{+2}_{-5}\%$), would be 109 ± 4 fs. This result is excluded by all previous measurements of the lifetime (see Table 27.8 in Endt [9]). Similarly, the inferred ground state branching ratio from the present results, assuming the literature values for spin ($J = 9/2$) and lifetime (85 ± 4 fs), would be $78 \pm 3\%$. This value is excluded by the measured upper limits on possible branching to all other levels. Joint variance of the lifetime and the branching ratio within accepted literature values also cannot explain the present integrated cross section, as evident in the uncertainty for the determined integrated cross section assuming a spin of $J = 9/2$ shown in Table I. Given the substantial prior measurements of the properties of this level, and the uncertainty of the spin of the level, a more likely explanation for the present integrated cross section is for a spin reassignment of $J = (7/2)$, but we cannot rule out the possibility that previous measurements of the lifetime or branching ratio are incorrect; additional measurements would be needed to explore that possibility.

The present suggestion for a spin reassignment to $J = (7/2)$ for the 3.004-MeV level would confirm the predictions of the original (though antiquated) strong-coupling (Nilsson) model. This result is, however, in conflict with the more-recent shell model calculations using the "universal" s-d interaction [10]. If an experiment were to establish the parity of the state as negative, which would be in contradiction with the supported parity determined from most previous experiments, then the state could be interpreted as an intruder state from the fp shell which was not included in the above shell model calculation.

A side effect of the present suggestion is that the spin of the $T = 1/2$ mirror partner in ^{27}Si of the presently considered level may also need to be reconsidered as $J = (7/2)$ [9]. The ^{27}Si 2.909-MeV level's present spin assignment of $J = 9/2$ is originally based on the mirror symmetry with the 3.004-MeV level in ^{27}Al, and an argument that angular momentum transfer to the levels are the same using similar reactions [25]. To fully resolve the issue, an additional measurement to determine the spin of the ^{27}Si 2.909-MeV level may be needed.

The present method of validating evaluated properties of a level by comparing its measured and expected NRF integrated cross section could help resolve questions on properties of levels in other nuclei. A spin determination may even be possible if the considered level has a reasonably well-known branching ratio and lifetime which enables the integrated cross section to be directly correlated to the spin, and a reasonably large ground state width to make the NRF measurement feasible. For example, the spins of the doublet in ^{31}P with energies of 5.0149 and 5.0152 MeV could be determined this way as both levels satisfy the above requirements. They currently have tentative spin values of $J^{\pi} = (3/2^{+})$ and $J^{\pi} = (1/2,3/2)^{-}$ [26]. Given the ground state spin of $J^{\pi} = 1/2^{+}$, the integrated cross sections for exciting a state with $J = 3/2$ will be double that of one having $J = 1/2$ allowing an easy distinguishment. Similarly, the spin of the 2.440-MeV state in ^{41}K with current tentative assignment of $J^{\pi} = (3/2,5/2^{+})$ [27] could be determined. As the ground state has a spin of $J^{\pi} = 3/2^{+}$, the difference in integrated cross section between the two values would be 50%.

IV. SUMMARY

In summary, we demonstrated how NRF can be used to validate evaluated resonance data by measuring the integrated cross section of the 3.004-MeV level in ^{27}Al, and comparing that result with the expected integrated cross section calculated using the accepted evaluated values for the spin, lifetime, and ground state branching ratio of the level. In the present case, we measured the integrated cross section to be 3.30 ± 0.10 (errors added in quadrature). This is an order of magnitude improvement in uncertainty over previous measurements. This value was found to be inconsistent with that expected from the evaluated properties, particularly using the tentative spin assignment of $J = (9/2)$. Consistency can be brought about if a spin of $J = 7/2$ is used. Modifying the spin to achieve consistency is preferred as it has not been firmly established, and because the branching ratio and lifetime of the 3.004-MeV level were well determined by previous measurements using other techniques. This result suggests an additional measurement to verify the spin of the 3.004-MeV level is warranted. A side effect of the present suggestion for a spin reassignment of the 3.004-MeV level is that a spin reassignment of the 2.909-MeV $T = 1/2$ mirror state in ^{27}Si to $J = (7/2)$ may be warranted. The present method has a unique advantage in that it is simpler and faster than other methods (particularly charged particle induced reactions) typically used to directly measure the spin or lifetime of a level.

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