Activation of $^{115}In^m$ by single pulses of intense bremsstrahlung

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A new technique has been recently described for the absolute calibration of intense sources of pulsed radiation in the 0.2–1.5 MeV range of photon energies. An activation technique, it depended upon the storage of samples of the irradiating spectrum in the form of populations of nuclei excited to isomeric states with lifetimes of seconds to hours. Described here is the use of such a calibrated source to resolve severe conflicts in previous studies of the reaction $^{115}In(\gamma,\gamma')^{115}In^m$ through the 1078 keV $J^\pi=\frac{5}{2}^+$ level; this mode has traditionally served as the archetype for (γ,γ') reactions. We report an integrated cross section of $(18.7\pm2.7)\times10^{-29}$ cm^2 keV with no evidence of any importance of nonresonant channels of excitation.

I. INTRODUCTION

For the study of (γ,γ') reactions that produce isomeric products, ^{115}In has a particularly favorable combination of characteristic properties. Having only a few channels for reaction at energies below 1.4 MeV, it nevertheless displays a large integrated cross section for excitation of the 269 min isomer at 336 keV. For these pragmatic reasons ^{115}In has served as the archetype material for the study of this type of reaction and a number of efforts have been reported[1–13] in the past 48 years.

The relevant part of the energy level diagram[14] of ^{115}In is shown in Fig. 1, indicating only three levels through which a (γ,γ') reaction of multipolarity $E1$, $M1$, or $E2$ could proceed to populate the isomeric state for photons below 1.4 MeV. The importance of the lowest gateway level at 941 keV is negligible because it has a particularly small integrated cross section[15] for excitation in comparison to that of the nearby 1078 keV level, and the 934 keV $\frac{7}{2}^+$ level has a yet smaller cross section because of its longer lifetime.

In practical cases in which ^{115}In samples are excited either with gamma rays from a source or by bremsstrahlung from an accelerator operating below 1.4 MeV, the absorption spectrum for (γ,γ') reactions producing isomers is thus essentially monochromatic at 1078 keV. Nevertheless, measurements by different investigators have shown considerable variance. Table I presents a summary of values reported in the literature together with the results of this measurement in terms of the integrated cross section as usually reported,[16] $\pi b_a b_0 \Gamma \sigma_0/2$, where Γ is the natural width in keV of the pump band, where the branching ratios b_a and b_0 give the probabilities for the decay of the gateway level back into the initial and isomeric level, respectively, and σ_0 is the Breit-Wigner cross section for the absorption transition,

$$\sigma_0=\frac{\lambda^2}{2\pi}\frac{2I_e+1}{2I_g+1}\frac{1}{\alpha_p+1}, \quad (1)$$

where λ is the wavelength in cm of the gamma ray at the resonant energy, E_i, I_e, and I_g are the nuclear spins of the excited and ground states, respectively, and α_p is the total internal conversion coefficient for the absorption transition. The α's can reasonably be expected to be less than 0.01 for the transitions discussed here.

It has been recently argued[16] that the principal cause of such a large degree of variance among previous measurements of the ^{115}In excitation has been the generally inadequate level of characterization of the spectrum of the pump source. It is particularly awkward to specify the spectrum from a line source. In a previous paper[13] we showed that the spectrum from a pulsed source of intense bremsstrahlung could be determined to a level of accuracy sufficient for the quantitative description of the reactions $^{77}Se(\gamma,\gamma')^{77}Se^m$ and $^{79}Br(\gamma,\gamma')^{79}Br^m$. This agreement now provides a reliable scheme to normalize other results to the pumping spectrum.

It is the purpose of this paper to report the reexamination of the reaction $^{115}In(\gamma,\gamma')^{115}In^m$ with the same pulsed bremsstrahlung source used for the reconciliation of the absorption cross sections to $^{79}Br^m$ and $^{77}Se^m$. The quantitative value for the integrated cross section we report is in good agreement with the value reported most recently as the result of excitation with a radioactive source.[12]

II. METHODS AND APPARATUS

In this method based on the results of Refs. 13 and 16, the uncertainty in the absolute value of the geometric coefficient coupling the source of pump radiation to the absorbing target is eliminated by normalizing both the

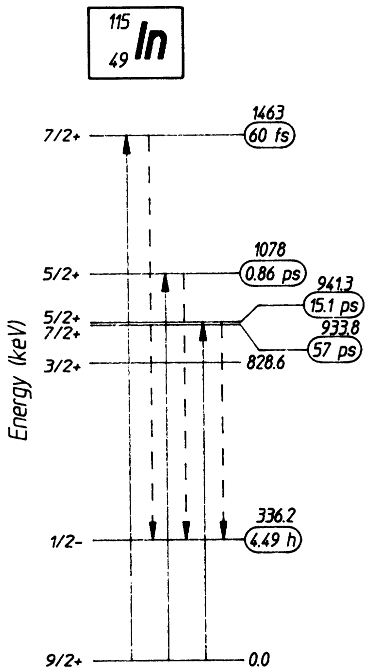

FIG. 1. Energy level diagram of the excited states of ^{115}In important in the production of populations of the isomer (Ref. 14). Half-lives of the states are shown to the right of each and sequences of (γ,γ') reactions leading to the isomer are shown by the arrows. Dashed γ' transitions occur by cascading through levels not shown.

pump fluence and the fluorescence counts to some standard material having a monochromatic excitation spectrum. The reaction $^{79}Br(\gamma,\gamma)^{79}Br^m$ is that standard, having an integrated cross section of 6.2×10^{-29} cm^2 keV and a convenient radioactivity in the isomer. Following the formalism reported earlier,[13] the number of $^{115}In^m$ nuclei, S(In), which could be excited by a flash of intense bremsstrahlung can be expressed as a ratio,

$$\frac{S(\mathrm{In})}{S(\mathrm{Br})}=\frac{N(\mathrm{In})}{N(\mathrm{Br})}\frac{\xi_{1078}(\mathrm{In})}{\xi_{761}(\mathrm{Br})}\zeta(1078)\ , \tag{2}$$

where $S(x)$ and $N(x)$ are the number of nuclei produced and the number of target nuclei of material x, respectively, $\zeta(1078)$ is the ratio of pumping intensity in units of (keV/keV) at 1078 keV to the intensity in (keV/keV) at 761 keV, and the $\xi_E(x)$ are the combinations of nuclear parameters involved in the excitation of the gateway level at energy E,

$$\xi_E(x)=\frac{(\pi b_a b_0 \Gamma \sigma_0/2)_E}{E}\ . \tag{3}$$

The collection of terms in parentheses in Eq. (3) comprises the integrated cross section for excitation as usually reported and the calibration value[13] for ^{79}Br is $\xi_{761}(\mathrm{Br})=8.2\times10^{-32}$ cm^2.

The source of excitation in these experiments[13] was the bremsstrahlung produced by the DNA PITHON nuclear simulator at Physics International. For these particular experiments the nominal firing parameters were deliberately perturbed so that successive irradiations could be obtained with end point energies verying from 0.9 to 1.5 MeV.

Intensities at the target were determined by measuring the nuclear activation of the ^{79}Br component of a sample of LiBr containing isotopes in natural abundance. This calibrating target was run in a pneumatic transfer system which enabled the population of $^{79}Br^m$ produced by a single irradiation to be subsequently counted with a NaI(*Tl*) detector of known efficiency at a quiet location 30 m removed from the source. Activation lost during the 1.0 s transit time could be readily corrected during analysis.

The ^{115}In sample under study was in the form of a thin foil taped to a fiduciary mark near the pneumatic system. Since the $^{115}In^m$ had a substantially longer half-life, it could be manually detached after exposure and transferred to the spectrometer which consisted of a 3"×3" NaI(*Tl*) detector with associated electronics. In typical cases a counting time of 1 h gave better than 2%

TABLE I. Summary of integrated cross sections reported for the reaction $^{115}In(\gamma,\gamma')^{115}In^m$ through the 1078 keV $J^\pi=\frac{5}{2}^+$ level.

Cross Section $\pi b_a b_0 \Gamma \sigma_0/2$ (10^{-29} cm^2 keV)	Method[a]	Reference
23±4	*S*	Ikeda and Yoshihara (Ref. 4)
20±4	*S*	Veres (Ref. 5)
7.1±2.3	*A*	Chertok and Booth (Ref. 6)
11.5±4.0	*A*	Booth and Brownson (Ref. 7)
30(+40, −20)	*A*	Boivin, Cauchois, and Heno (Ref. 8)
10.5±2.7	*S*	Lakosi, Csuros, and Veres (Ref. 9)
19±1	*S*	Watanabe and Mukoyama (Ref. 10)
5.39±0.64	*S*	Ljubici, Pisk, and Logan (Ref. 11)
18.1±1.5	*S*	Yoshihara *et al.* (Ref. 12)
14±1		Calculated from lifetimes and branching ratios (Ref. 14)
18.7±2.7	*A*	This work

[a] *A* is bremsstrahlung from an accelerator; *S* is radiation from ^{60}Co source.

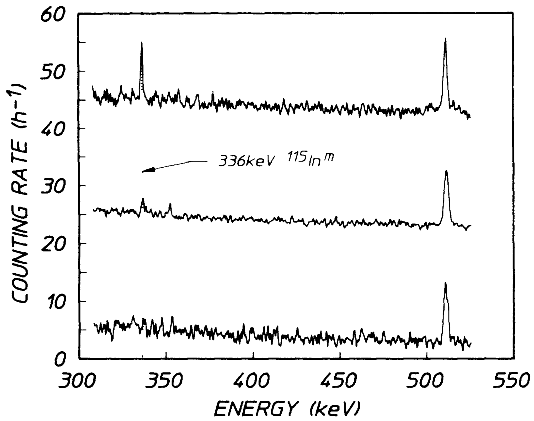

FIG. 2. Three sequential spectra from an intrinsic Ge detector begun at times 6.5, 9.2, and 19.0 h after the irradiation with a flash of bremsstrahlung with an 1.3 MeV end point. Data have been offset by 40, 20, and 0 counts/h, respectively. The 336.2 keV peak is seen to decay with the appropriate half-life of 4.49 h for $^{115}In^m$. The other structure is the annihilation peak at 511 keV, present in the background at a constant rate.

typical cases a counting time of 1 h gave better than 2% statistical accuracy in the $^{115}In^m$ peak after removal of background. In the course of this experimental series 12 shots were obtained for sufficiently high end point energies to yield fluorescent isomeric activity for ^{115}In.

To confirm that the fluorescence being detected resulted only from decay of the $^{115}In^m$ activity, additional foils were irradiated and then examined with an intrinsic Ge spectrometer, carefully shielded. In Fig. 2 we show spectra from an In foil irradiated by bremsstrahlung with an endpoint of 1.3 MeV. The fluorescence peak is at 336.2 keV, with a half-life seen to be consistent with a tabulated value of 4.49 h.

The relative bremsstrahlung intensity emitted at 1078 keV was a strong function of end-point energy of the accelerator as shown in Fig. 3. These data were obtained

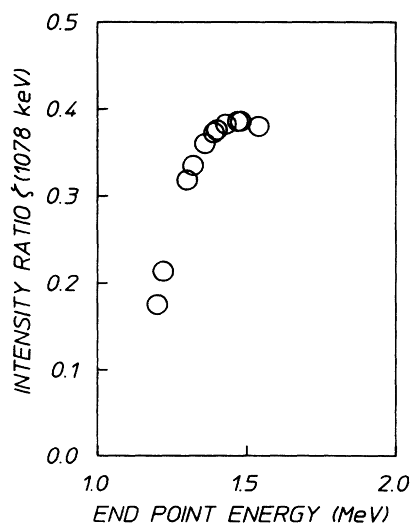

FIG. 3. Plot of the ratio of intensities of the bremsstrahlung spectrum at 1078 keV to that at 761 keV as a function of the end-point energy of the electron beam producing the photons. Statistical uncertainties are smaller than the data points.

by numerically fitting theoretical computations of bremsstrahlung spectra. A sample computed spectrum and the verification points from ^{79}Br and ^{77}Se are shown in Ref. 13. For each shot of the PITHON simulator the voltage and current were integrated in a bremsstrahlung modeling code to normalize the activity induced. As described previously,[13] confidence in the present method was established by examining quantitatively the number of fluorescent nuclei produced by successive irradiations of samples of ^{79}Br and ^{77}Se at a variety of end-point energies. The monotonic results in Fig. 3 indicate that the end point determination is reliable to within ±50 keV from shot to shot.

III. RESULTS

Because of a physical displacement between the ^{115}In sample and the mixed $^{79}Br/^{77}Se$ calibrator, the observed activities required correction for the different x-ray intensity obtained at these two points. This was done by mounting thermoluminescent dosimeters (TLD's) at both positions and comparing delivered doses for each shot. Using the assumption that the relative spectral distribution was constant even though the intensity was varying with position, the ^{115}In activity was scaled by the ratio of the calibrator dose to that of the indium sample. This correction ranged from 1.40 to 0.69 for the shots used in this work. The relative reversals of the positions of calibrator and sample with respect to the center of the irradiation source gave the same results, thus supporting the validity of the method of correction.

The indium sample was optically thin (0.183 g/cm^2) at the 336 keV energy of the fluorescence from the $^{115}In^m$ thus obviating corrections for self-absorption. Data were corrected for fluorescence and detector efficiencies. The resulting values of $S(\mathrm{In})/S(\mathrm{Br})$ are obtained from Fig. 4 by multiplying the ratios of fluorescent counts shown there by 0.75, the ratio of correction factors for these effects.

The linear form of the dependence of the relative yield of fluorescence from the indium isomers seen in Fig. 4 between 1.0 and 1.45 MeV is a strong indication of the dominance of a single channel of excitation through a gateway level lying at an energy given by the value of intercept. From the data of Fig. 4 it is seen that the intercept lies between 1000 and 1200 keV in agreement with the known $\frac{5}{2}^+$ state at 1078 keV. Also indicated in Fig. 4 is the energy of the next higher gateway state, $\frac{7}{2}^+$ at 1463 keV. It is interesting to observe that for end-point energies above this value there may be a tendency of the data to depart from the simple linear fit because of the availability of this additional gateway. A greater number of measurements at successively higher end-point energies would be needed to confirm this indication.

Once the data below 1.4 MeV are established as being consistent with the model of excitation through a single level at 1078 keV, Eq. (2) provides a means of determining the absolute cross section for the excitation. In Fig. 5 data for the measured values on the left side of Eq. (2) are plotted as functions of the relative intensities, $\zeta(1078)$, appearing on the right.[17] The scatter in the counting ra-

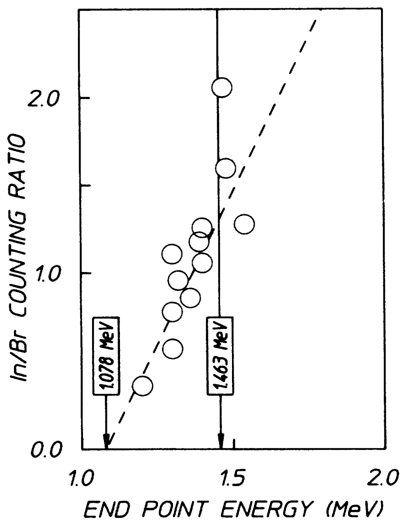

FIG. 4. Ratios of fluorescent photons from $^{115}In^m$ to those from $^{79}Br^m$, produced by single discharges from PITHON, as corrected for the finite duration of the counting interval and plotted as a function of the end-point energies of the electrons producing the bremsstrahlung. The dashed line shows a linear fit to the data intercepting the x axis at a single gateway energy of 1.078 MeV. The excitation energy of the next higher gateway is also shown at 1.463 MeV. Statistical uncertainities are smaller than the data points.

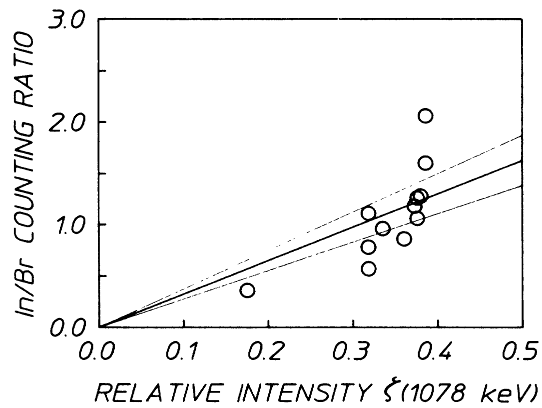

FIG. 5. Plot of the ratios of $^{115}In^m$ to $^{79}Br^m$ fluorescence observed as a function of the relative intensity of irradiation at 1078 keV normalized to the intensity at 761 kev. The heavy line shows the least-squares fit including the origin and the lighter lines indicate the uncertainty in such a fit introduced by varying the characterization of the intensities of Fig. 3 over the uncertainties in the characterization of the source. Slopes must be multiplied by 0.75, the product of the ratio of corrections for detector and fluorescence efficiencies and for self-absorptions in the targets before used in expressions such as Eq. (2).

tio is not due to the 2% counting statistics, but to the scatter in the modeled normalized fluxes, for which consistency provides the best estimate of the uncertainty. Note that it is the shape, not the absolute intensity of the pumping spectrum that enters as $\xi(1078)$. As can be seen from Eq. (2), the best slope around which the data of Fig. 5 scatter would represent our experimental determination of the product of the first two terms on the right of Eq. (2).

From the linear fit to the data of Fig. 5 shown as the heavy line, Eqs. (2) and (3), and the value of integrated cross section for ^{79}Br mentioned earlier, we obtain $\xi_{1078}(\mathrm{In})=(17.3\pm2.5)\times10^{-32}$ cm^2. The uncertainty was obtained from application of the same analyses to the lighter lines in Fig. 5 bounding the scatter from the least-squares fit to the data. The lines shown were obtained by determining the extent to which the fit to the data points of Fig. 5 would be displaced horizontally if the corresponding abscissae were varied by the maximum extent of the uncertainty in the value of $\xi(1078)$ arising from the nature of the calibration and the interpolations being employed,[13] that is, by the uncertainty in the slope of the pumping spectrum. The scatter of the data is within the bounds established by this uncertainty in intensity. Not explicitly shown in $\xi_{1078}(\mathrm{In})$ is the $\pm16\%$ uncertainty in the reference value of $\xi_{761}(\mathrm{Br})$ taken from the literature.[14]

Finally, substituting the result for $\xi_{1078}(\mathrm{In})$ into Eq. (3) and solving for the integrated cross section gives for the 1078 keV transition in ^{115}In, $\pi b_a b_0 \Gamma \sigma_0/2=(18.7\pm2.7)\times10^{-29}$ cm^2 keV. This is the value we report in Table I.

IV. CONCLUSIONS

The detailed characterization of the spectrum emitted by the intense source of pulsed bremsstrahlung described earlier[13] has been found to be sufficient to determine the yield of the reaction $^{115}\mathrm{In}(\gamma,\gamma')^{115}\mathrm{In}^m$. Table I shows this value to agree with some of the prior measurements. In this work there was no need to invoke any nonresonant reaction channels of the type sometimes used[11] in the description of this reaction.

In addition to providing further evidence against the occurrence of nonresonant reactions in ^{115}In, the results of this work have important implications for the calibration of intense sources of pulsed continua. By providing a means for storing a sample of the illuminating intensity at a single well-defined energy of 1078 keV for subsequent measurement at a later, quieter time, a sample of ^{115}In can readily complement the information supplied[13,16] by ^{79}Br about the intensity at 761 keV. Moreover, both together can be used to identify the component of excitation contributed by the higher energy lines of ^{77}Se so that the remainder can be used to characterize the intensities at lower energies.[13]

Finally, it seems this technique of using single pulses of intense continua to measure integrated cross sections for the production of measurable populations of isomers can provide data of use in astrophysical modeling. Cross section at photon energies as low as 1 MeV are quite large for such elements as Se, Br, and In and might provide viable photonuclear channels for the production of enough isomeric population to be important in cosmic nucleosynthesis. It seems that the experiments reported here give evidence that it is possible to advance the precision for the characterization of (γ,γ') reactions toward that enjoyed by other types of particle reactions at comparable energies.

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