

THE DECAY OF ^{75}Se D. E. RAESIDE, M. A. LUDINGTON, J. J. REIDY and M. L. WIEDENBECK
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Abstract: The gamma-ray spectrum associated with the electron capture decay of ^{75}Se has been studied with Ge(Li) spectrometers. Coincidence spectra were taken with a Ge(Li)-Ge(Li) combination. The data do not support a recent report of several new gamma rays in the decay of ^{75}Se . Gamma-gamma angular correlation experiments were performed with a NaI(Tl)-Ge(Li) combination. The value obtained for $\delta(280\text{ keV})$ is in excellent agreement with one recently reported value but in poor agreement with another. The ratio P_K/P_{tot} for the electron capture branch to the 401 keV level of ^{75}As has been determined, and the result agrees with previous experimental determinations.

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RADIOACTIVITY ^{75}Se ; measured E_γ , I_γ , $\gamma\gamma$ -coin., γX -coin., $\gamma\gamma(\theta)$;
deduced K-capture fraction P_K/P_{tot} . ^{75}As deduced levels.

1. Introduction

The electron capture decay of ^{75}Se to levels in ^{75}As has been studied extensively. A compilation and evaluation of the experimental data available prior to 1967 have been made by Pancholi and Ikegami of the Nuclear Data Group¹). With the advent of large volume Ge(Li) detectors, the opportunity to obtain more accurate gamma-ray intensities and improved coincidence results presented itself. Of special interest was the opportunity to make use of Ge(Li) detectors in re-investigating the angular correlations between the ^{75}As gamma rays.

2. Gamma-ray energy and intensity measurements

2.1. EXPERIMENTAL ARRANGEMENTS

A Nuclear Diodes 40 cm³ trapezoidal Ge(Li) detector was used for the measurement of gamma-ray energies and intensities. The associated electronics consisted of a Tennelec TC135 pre-amplifier, a Tennelec TC200 amplifier and a Scipp 1600-channel analyser. The resolution of the system was 2.5 keV at 662 keV. An energy calibration for gamma rays between 60 keV and 401 keV was obtained by performing a least-squares fit to the energies and channel positions of four gamma rays, e.g. ^{109}Cd (88.032 ± 0.003 keV), ^{139}Ce (165.854 ± 0.011 keV), ^{22}Na (511.006 ± 0.002 keV) and ^{137}Cs (661.635 ± 0.076 keV). The 265 keV, 280 keV, 304 keV and 401 keV gamma rays of ^{75}Se provided the four calibration points utilized in the measurement of the ener-

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gies of the 419 keV, 572 keV and 617 keV gamma rays. The non-linearities in the system were accounted for using the method of Donnelly *et al.* ²⁾. A relative efficiency calibration ²⁾ for the system was obtained for the range 90 keV to 1400 keV by utilizing sources for which the relative emission rates of pairs of gamma rays are precisely known, e.g. ^{180m}Hf, ^{108m}Ag, ²²Na, ⁶⁰Co and ⁴⁶Sc. For gamma-ray intensities in the energy range below 90 keV, a modified pair-point method ³⁾ was used. In this method, a comparison is made of the relative peak areas of the gamma ray de-exciting a single-level and the X-rays which follow internal conversion. By using seven sources (¹⁰⁹Cd, ¹³⁷Cs, ¹³⁹Ce, ¹⁶⁰Tb, ¹⁷⁷Lu, ¹⁵⁵Eu and ⁵⁷Co), the relative efficiency calibration was extended to 20 keV.

Source material was obtained from New England Nuclear Corporation in the form of a dilute HCl solution. The source was prepared by allowing a small drop of this solution to dry on a lucite disk.

2.2. RESULTS

The results of the gamma-ray energy and intensity measurements are presented in table 1. Two of the Ge(Li) spectra used for these measurements are shown in figs. 1 and 2. There is no evidence in these spectra for gamma rays peaks at 335 keV, 415 keV

TABLE 1
Energies and intensities of gamma rays in the decay ⁷⁵Se $\xrightarrow{E.C.}$ ⁷⁵As

Energy (keV)	Relative gamma-ray intensity
65.98 ± 0.10	1.40 ± 0.40
96.71 ± 0.07	4.83 ± 0.96
121.10 ± 0.07	29.2 ± 2.9
136.01 ± 0.05	96.0 ± 9.6
198.71 ± 0.09	2.25 ± 0.23
264.68 ± 0.06	100
279.53 ± 0.06	41.3 ± 4.1
303.99 ± 0.08	2.06 ± 0.21
400.75 ± 0.06	19.2 ± 1.9
419.15 ± 0.30	0.020 ± 0.003
572.24 ± 0.34	0.053 ± 0.008
617.48 ± 0.43	0.0076 ± 0.0010

or 486 keV. Gamma rays of this energy were recently reported by Nagpal ⁴⁾. The peaks which Nagpal observed in the energy range between 525 keV and 560 keV appear to be due to summing. To demonstrate this we ran several spectra at distances varying from 1 cm to 20 cm (see fig. 2). Intensity upper limits (relative to the intensity of the 265 keV gamma ray, $I_\gamma(265 \text{ keV}) \equiv 100$) for some of the gamma rays reported by Nagpal are $I_\gamma(335 \text{ keV}) < 2 \times 10^{-2}$, $I_\gamma(486 \text{ keV}) < 10^{-3}$, $I_\gamma(529 \text{ keV}) < 2 \times 10^{-3}$, $I_\gamma(536 \text{ keV}) < 2 \times 10^{-3}$, $I_\gamma(543 \text{ keV}) < 2 \times 10^{-3}$ and $I_\gamma(556 \text{ keV}) < 10^{-3}$.

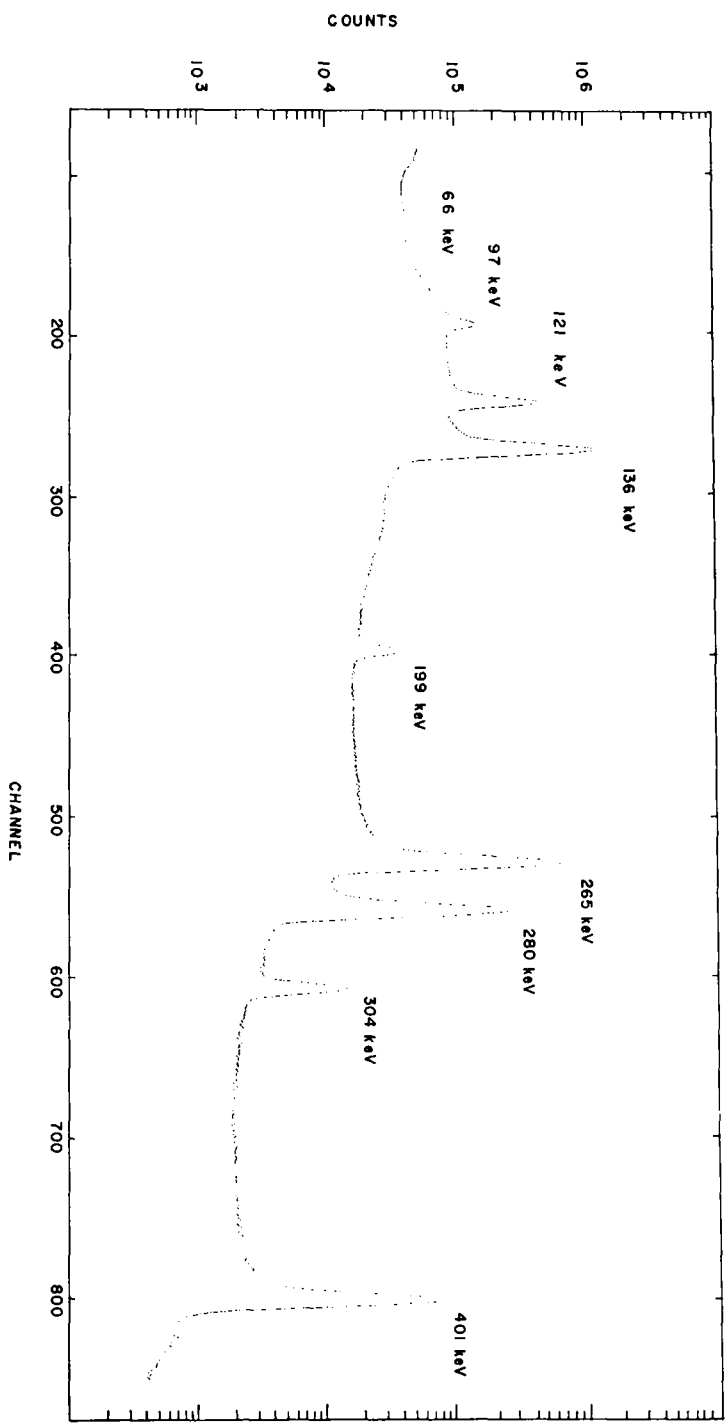


Fig. 1. Gamma-ray spectrum of ^{75}Se from 25 keV to 420 keV observed with a 40 cm^3 Ge(Li) spectrometer.

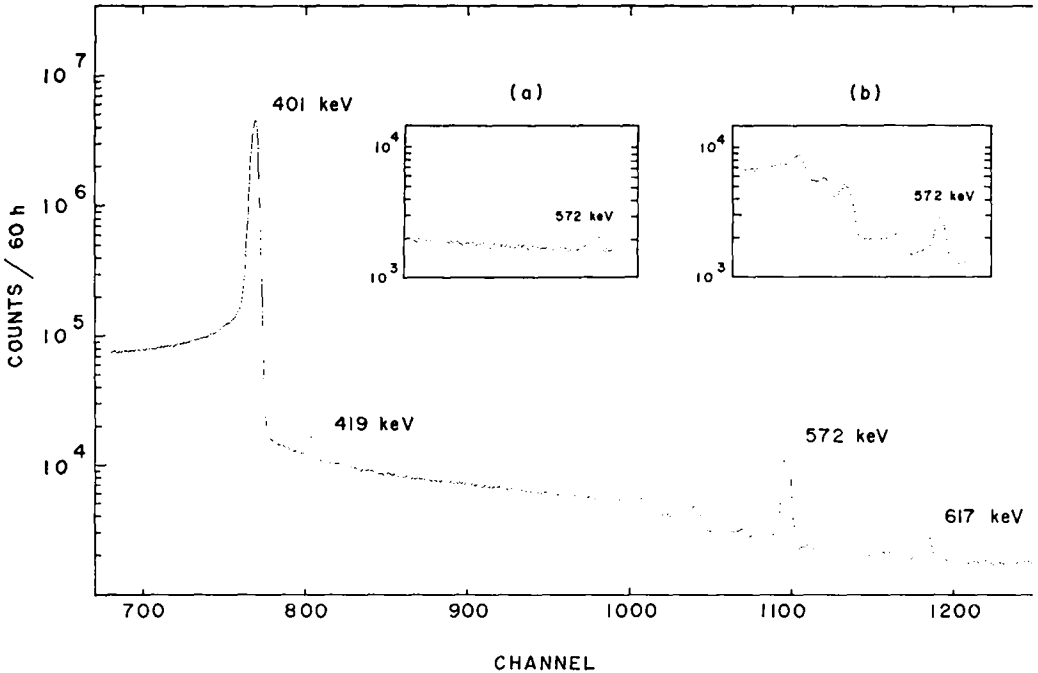


Fig. 2. Gamma-ray spectrum of ^{75}Se from 350 keV to 660 keV observed with a 40 cm^3 Ge(Li) spectrometer. The data shown in the main spectrum were obtained in a run utilizing lead absorbers at a source-to-detector distance of 5 cm. Inset (a) shows data taken at 20 cm (with absorbers) and inset (b) shows data taken at 5 cm (without absorbers).

3. Gamma-gamma coincidence studies

3.1. EXPERIMENTAL ARRANGEMENTS

The source used for the measurement of gamma-ray energies and intensities was also used for the gamma-gamma coincidence study. The total spectrum channel of the coincidence system consisted of an Ortec 32 cm^3 coaxial Ge(Li) detector coupled to a Canberra 1408C pre-amplifier and Tennelec TC200 amplifier. The gate channel consisted of a Nuclear Diodes 40 cm^3 trapezoidal Ge(Li) detector coupled to a Nuclear Diodes 101 pre-amplifier, a Tennelec TC200 amplifier and TC250 biased amplifier and stretcher. The resolution of both detectors was about 2.5 keV at 662 keV. The coincidence electronics was of the fast-slow type. Leading-edge timing was used for both channels. A resolving time 2τ of 125 ns was used for all runs. Coincidence spectra were displayed on a Scipp 1600-channel analyser.

3.2. RESULTS

A partial summary of the results of the gamma-gamma coincidence study is shown in table 2. A level scheme which is consistent with the results of the present investi-

TABLE 2
Gamma-gamma coincidence results

Gate (keV)	Coincident gamma ray ^{a)} (keV)
121	280
136	66, 199, 265
199	66, 136, 420
265	136
280	121
401	none

^{a)} The gamma rays indicated were those observed to be in coincidence with the gated gamma ray after chance subtraction (2τ determined with a time-to-pulse-height converter) and allowance for coincidences due to Compton scattered photons in the gate (regions adjacent to peaks gated separately).

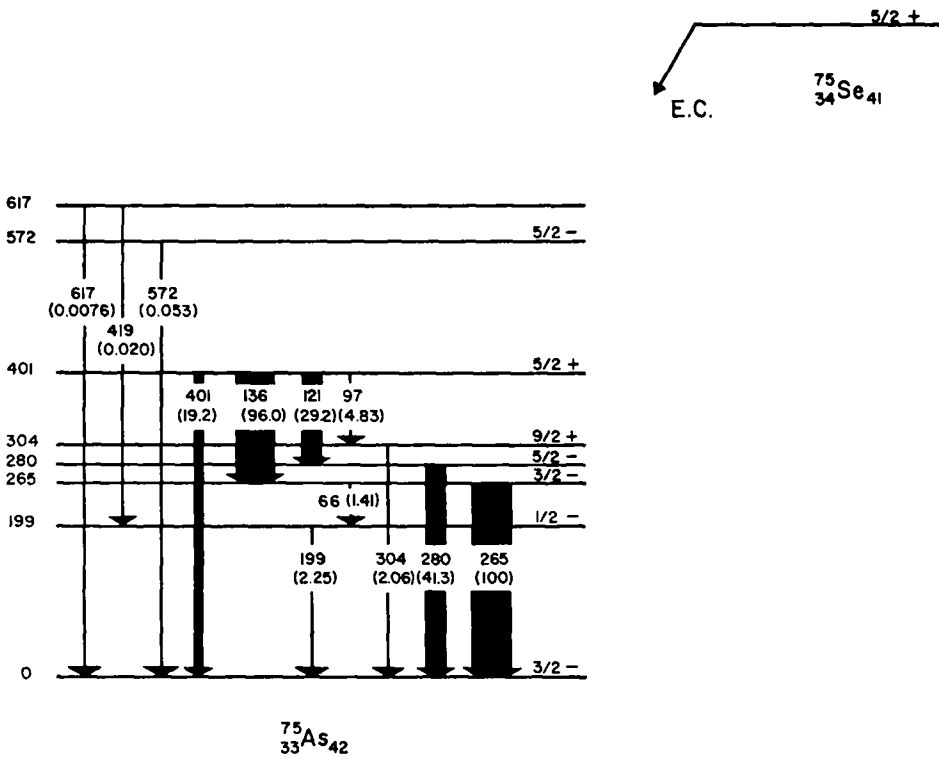


Fig. 3. The level scheme of ⁷⁵As populated in the decay of ⁷⁵Se. Spins and parities are from ref. ¹⁾.

gation is shown in fig. 3. This level scheme is in agreement with the work of previous investigators ¹⁾.

4. Gamma-gamma correlation measurements

4.1. EXPERIMENTAL ARRANGEMENTS AND MEASUREMENT METHOD

The source material used for the gamma-gamma angular correlation measurements was taken from the same solution utilized in the energy, intensity and gamma-gamma coincidence measurements. A drop of the solution was placed in the bottom of a thin-walled lucite cylinder. One detection channel consisted of a planar $8 \text{ cm}^2 \times 5 \text{ mm}$ Ortec Ge(Li) detector coupled to an Ortec 118A pre-amplifier, the other consisted of a Harshaw $7.6 \text{ cm} \times 7.6 \text{ cm}$ NaI(Tl) crystal (integral mount) coupled to a Hamner N-381 pre-amplifier. The coincidence electronics was of the fast type and used cross-over timing in both channels. The singles counts in each channel and the total coincidence count were recorded on Sodeco print-out units. Data were taken automatically in a double quadrant sequence; the data were collected at seven equally spaced angles in each quadrant with print-out occurring after ten minutes accumulation of counts. The resolving time was determined between runs by placing a $0.5 \mu\text{s}$ delay in one channel with respect to the other and collecting the chance coincidences. The resolving time 2τ was 65 ns, which gave a true-to-chance ratio of 6 : 1.

Data were analysed by computer. The computer program provided for a chance subtraction for each coincidence print-out, division of the resulting true coincidence count by the product of the singles counts and the folding together of these normalized values into a set representing the true coincidences associated with the seven angles 90° , 105° , 120° , 135° , 150° , 165° and 180° . The computer performed a least-squares fit of this set to the function $W(\theta) = A_0 + A_2P_2(\cos \theta) + A_4P_4(\cos \theta)$ following the method outlined by Rose ⁵). The ratios A_2/A_0 and A_4/A_0 and the uncertainties associated with these ratios ⁶) constituted the computer output. Geometric corrections for the NaI(Tl) crystal were obtained from the computed values of Herskind and Yoshizawa ⁷), while the geometric corrections for the Ge(Li) crystal were experimentally determined by means of a collimated-beam experiment. Since both crystals subtended solid angles of approximately 1 %, these corrections were small.

4.2. RESULTS

The 136-265 keV correlation. This angular correlation was obtained by gating the 136 keV peak in the Ge(Li) channel and the unresolved 265, 280 keV peak in the NaI(Tl) channel. After making the necessary geometric corrections, we obtain

$$(A_2/A_0)_{136-265} = -0.033 \pm 0.004,$$

$$(A_4/A_0)_{136-265} = 0.001 \pm 0.009.$$

These ratios were obtained from a least-squares fit which utilized more than 10^7 true coincidences at each of the seven angles to which the fit was made.

The 121-280 keV correlation. This angular correlation was obtained by gating the 121 keV peak in the Ge(Li) channel and the unresolved 265, 280 keV peak in the

NaI(Tl) channel. After making the necessary geometric corrections, we obtain

$$(A_2/A_0)_{121-280} = -0.404 \pm 0.006,$$

$$(A_4/A_0)_{121-280} = 0.002 \pm 0.009.$$

These ratios were obtained from a least-squares fit which utilized more than 10⁶ true coincidences at each of the seven angles to which the fit was made.

TABLE 3
Comparison with previous measurements of the (121-280 keV) and (136-265 keV) gamma-gamma angular correlations

Ref.	Detector	Cascade	A ₂ /A ₀	A ₄ /A ₀
9)	NaI(Tl)-NaI(Tl)	121-280	-0.42 ± 0.03 ^{a)}	-0.014 ± 0.017
		136-265	-0.019 ^{+0.01} _{-0.02}	-0.012 ± 0.012
10)	NaI(Tl)-NaI(Tl)	121-280	-0.41 ± 0.03	
		136-265	0.016 ± 0.03	
11)	NaI(Tl)-NaI(Tl)	121-280	-0.465 ± 0.016 ^{b)}	
		136-265	-0.011 ± 0.009 ^{b)}	
12)	NaI(Tl)-Ge(Li)	121-280	-0.430 ± 0.006	0.009 ± 0.007
		136-265	-0.033 ± 0.001	0.001 ± 0.002
13)	Ge(Li)-Ge(Li)	121-280	-0.404 ± 0.004	-0.009 ± 0.008
		136-265	-0.0302 ± 0.0029	0.0036 ± 0.0056
this work	NaI(Tl)-Ge(Li)	121-280	-0.404 ± 0.006	0.002 ± 0.009
		136-265	-0.033 ± 0.004	0.001 ± 0.009

^{a)} The average of two values given in ref. 9).

^{b)} Calculated from the coefficient of cos² θ given in ref. 11).

Both the (121-265 keV) and (136-280 keV) cascades proceed through intermediate states having mean lifetimes ⁸⁾ shorter than 4 × 10⁻¹⁰ sec; therefore, we expect no attenuation of the directional correlation associated with either cascade. No corrections were needed for interfering correlations.

Table 3 shows a comparison between the angular correlation results of this study and those of previous investigators.

4.3. THEORETICAL CONSIDERATIONS

The data of the present work support the previously suggested ¹¹⁻¹³⁾ spin sequences and multiplicities for the (121-280 keV) and (136-265 keV) cascades

$$\frac{3}{2}(D)\frac{3}{2}(D, Q)\frac{3}{2} \quad (121-280 \text{ keV}),$$

$$\frac{3}{2}(D)\frac{3}{2}(D, Q)\frac{3}{2} \quad (136-265 \text{ keV}).$$

Conversion coefficient data ¹⁾ show that both the 121 keV and 136 keV transitions are pure dipole. We calculate the following mixing ratios for the 265 keV and 280 keV transitions:

$$\delta(265) = -0.045 \pm 0.025 \text{ or } 4.7^{+0.7}_{-0.5},$$

$$\delta(280) = -0.393^{+0.015}_{-0.017} \text{ or } -1.28^{+0.04}_{-0.03}$$

Only the first value listed for each mixing ratio is compatible with conversion coefficient data ^{12,13}). Our value for $\delta(280 \text{ keV})$ is in agreement with the one given by Becker and Steffen ¹³) but does not agree with the one given by Speidel *et al.* ¹²).

5. The K-capture probability to the 401 keV level

5.1. EXPERIMENTAL ARRANGEMENTS

The source material used for the measurement of the K-capture probability to the 401 keV level of ⁷⁵As was taken from the same solution utilized in the energy, intensity, gamma-gamma coincidence and angular correlation measurements. A small drop of the solution was placed on a piece of mylar having a thickness of 0.000635 cm and allowed to dry. A second piece of the same mylar was then used to cover the source. This source assembly was mounted at the end of a cardboard tube which was in turn mounted on a Harshaw NaI(Tl) low noise crystal-photomultiplier assembly. The X-ray detector was a cleaved crystal having a diameter of 2.46 cm and a thickness of 0.0794 cm integrally mounted on an Amperex XP1010 photomultiplier tube. The detector had a 0.0127 cm thick beryllium window. The remaining components of the experimental apparatus used in the K-capture measurement were the 32 cm³ Ge(Li) detector and electronics utilized in the gamma-gamma coincidence part of the present study. A graded absorber was placed in front of the Ge(Li) detector to minimize the occurrence of accidental summing.

5.2. MEASUREMENT METHOD

The ratio of the probability of the occurrence of K-capture populating the 401 keV level to the probability of the occurrence of any form of electron capture to that level is given by

$$\frac{P_K}{P_{\text{tot}}} = \frac{N_{K-401}}{N_{401} T_K \epsilon_K \Omega_K \omega_K},$$

where N_{K-401} is the number of coincidences between the As K X-rays and the 401 keV gamma rays per unit time, N_{401} the number of 401 keV gamma rays per unit time, T_K the fraction of the K X-rays which pass unattenuated through the mylar, air and beryllium, ϵ_K the efficiency of the NaI(Tl) crystal for the 10.5 keV X-rays, Ω_K the fraction of the solid angle subtended by the source at the face of the NaI(Tl) crystal and ω_K the arsenic fluorescence yield for the K-shell. Gorski has calculated ¹⁴) that the efficiency of NaI(Tl) crystals having a thickness of 0.1 cm for photons of energy less than about 19 keV is unity, and that for crystals with a thickness of 0.2 cm the efficiency is unity for photons having an energy less than about 22 keV. This would indicate that we can safely assume that crystals having a thickness of 0.08 cm have unity efficiency for 10.5 keV photons.

The semi-empirical formula of Burhop ¹⁵)

$$\left(\frac{\omega_K}{1-\omega_K} \right)^{\frac{1}{2}} = A + BZ + CZ^3,$$

which relates the K-shell fluorescence yield ω_K to the atomic number Z , was used to determine the arsenic K-shell fluorescence yield. The most recent evaluation of the constants A , B and C is that of Hagedoorn and Wapstra ¹⁶). Since these constants are determined by a least-squares fit to a large number of experimental fluorescence yields, it seems preferable to accept the arsenic fluorescence yield value calculated from the Burhop formula rather than to make use of the one available experimental value ¹⁷) for the arsenic K-shell fluorescence yield. Using the constants $A = -6.4 \times 10^{-2}$, $B = 3.40 \times 10^{-2}$ and $C = -1.03 \times 10^{-6}$ determined by Hagedoorn and Wapstra ¹⁶), we calculate that $\omega_K(\text{As}) = 0.521$. The uncertainty to be associated with this value is 0.006 [ref. ¹⁶)]. The one experimental value for $\omega_K(\text{As})$, which was reported ¹⁷) without an estimate of uncertainty, is 0.53. Transmission correction factors for air and beryllium were obtained by graphical interpolation of the data compiled by Hubbell [ref. ¹⁸)]. A transmission factor for mylar was determined experimentally. Since the distance between the source and the NaI(Tl) crystal (11.1 cm) was large relative to the source diameter and the detector diameter, the geometric solid angle fraction was used, i.e. source size and edge effects were neglected.

The data were collected in a sequence of six runs with the resolving time 2τ being determined at the start and finish of each run by the two-source method. A resolving time of 205 ns was used for four runs. The true-to-chance ratio was 3 : 1 for these runs. The resolving time was increased to 365 ns for two runs. The same ratio N_{K-401}/N_{401} was obtained (within uncertainties) in these two runs as in the four runs at the smaller resolving time. This indicated that the coincidence efficiency was unity. More than 10000 true coincidences were collected in each of the six runs.

5.3. RESULTS

The weighted average of the ratio of the number of coincidences between the K X-rays and the 401 keV gamma rays per unit time to the number of 401 keV gamma rays per unit time is 0.00131 ± 0.00002 . Applying the necessary corrections to this ratio, we obtain $P_K/P_{\text{tot}} = 0.886 \pm 0.026$. An estimation of uncertainties associated with the correction factor has been included in this result. The value of P_K/P_{tot} reported by Rao *et al.* ¹⁹) is 0.885 ± 0.018 , where the value of $\omega_K(\text{As})$ was taken to be 0.520. Perrin ²⁰) reports a capture ratio based on $\omega_K(\text{As}) = 0.55$. His result is $P_K/P_{\text{tot}} = 0.83$ with no estimation of uncertainty given. The agreement between the three measurements is good when a common value for $\omega_K(\text{As})$ is adopted.

5.4. COMPARISON WITH THEORY

Theoretical calculations of electron capture ratios can be performed using the method given by Behrens and Jänecke ²¹). This method takes into account the effects of the finite size of the nucleus, the screening of the nuclear electrostatic field by the orbital electrons and electron exchange.

The following expression was used to obtain our theoretical estimate of P_K/P_{tot} for the electron capture branch to the 401 keV level of ^{75}As :

$$\frac{P_{\text{tot}}}{P_K} \approx 1 + \left(\frac{Q - B_{L_I}}{Q - B_K} \right)^2 \left(\frac{\beta_{L_I}}{\beta_K} \right)^2 \left[1 + \left(\frac{\beta_{L_{II}}}{\beta_{L_I}} \right)^2 \right] \left[1 + \left(\frac{Q - B_{M_I}}{Q - B_{L_I}} \right)^2 \left(\frac{\beta_{M_I}}{\beta_{L_I}} \right)^2 \right].$$

Numerical values of the squared Coulomb amplitudes β_K^2 , $\beta_{L_I}^2$, $\beta_{L_{II}}^2$ and $\beta_{M_I}^2$ were taken from the work of Behrens and Jänecke²¹). The electron binding energies B_K , B_{L_I} and B_{M_I} were taken from the data of the Uppsala Group which have been tabulated by Lederer *et al.*²²). The Q -energy available for K-capture to the 401 keV level in arsenic was calculated by subtracting 400.7 keV from the total energy available for the decay of ^{75}Se (864.6 keV) as given by the Nuclear Data Group²³). Upon substitution of numerical values, we obtain $(P_K/P_{\text{tot}})_{\text{B-J}} = 0.886$. This estimate is in excellent agreement with our experimental value.

Fink²⁴) has noted that there is now strong evidence in favor of discarding much of the old data used in previous fits to the Burhop formula. If this is the case, then our experimental value of P_K/P_{tot} might be changed by as much as several percent.

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