Nuclear Resonance Fluorescence in As^{75}†

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(Received December 12, 1957)

When As^{75} is irradiated with the gamma rays from a gaseous source of Se^{75}, strong resonance fluorescence from the 265-kev excited state is observed. The angular distribution of the resonance radiation has the form $W(\theta)=1+(0.15\pm0.02)P_2(\cos\theta)$, indicating that the spin of the 265-kev level is larger than $\frac{1}{2}$. If one assumes a spin value of $\frac{3}{2}$, the measured self-absorption of the resonance fluorescence leads to a mean life of $(1.6\pm0.2)\times10^{-11}$ second for the 265-kev transition to the ground state of As^{75}.

Resonance fluorescence from the 280-kev excited state was found to be at least one order of magnitude smaller than that from the 265-kev state. From this one concludes that the mean life of the 280-kev transition is longer than 10^{-10} second.

No resonance scattering from the 402-kev level was observed. On the basis of the source strength and of the geometry, the absence of an appreciable resonance effect establishes a lower limit of 10^{-10} second for the mean life of the 402-kev state, and of 7.10^{-10} second for the partial mean life of the 402-kev transition to the ground state.

INTRODUCTION

STUDIES[1] of the radioactive decay of Se^{75} established five excited states in As^{75} at excitation energies of 199, 265, 280, 305, and 402 kev. The 305-kev state was found[2] to be isomeric with a mean life of 17 milliseconds. Delayed coincidence experiments[1] showed that the 265- and 280-kev transitions have mean lives shorter than 1.1×10^{-9} second. A 199-kev transition and a line corresponding probably to a combination of 265- and 280-kev gamma rays have been observed in Coulomb excitation.[3] From the observed yields a partial $E2$ mean life of 6×10^{-9} second is calculated for the 199-kev transition while the yield at 280 kev, if attributed to a single line, corresponds to a mean life of $\sim10^{-9}$ second. Little is known about the 402-kev level which is deexcited by 97-, 121-, and 137-kev transitions competing successfully with the 402-kev $E1$ transition[2] to the ground state.

The short lifetime limits for the 265- and 280-kev transitions encouraged this study of As^{75} with the resonance fluorescence technique. Other favorable factors were the property of selenium to form many volatile compounds, the fact that arsenic is a monoisotopic element, the longevity of Se^{75} (127 days), and the low Z value of arsenic which promised little interference from nonresonant elastic scattering.

As far as the 199-, 280-, and 402-kev levels are concerned, the information obtained from the resonance experiments was fragmentary because the resonance effects were too small to be studied. The resonance fluorescence from the 265-kev level, on the other hand, was very strong indeed. For this level it was, therefore, possible to study, aside from the simple scattering, the self-absorption of the resonance radiation and its angular distribution.

GENERAL CONSIDERATIONS

The equations necessary for the analysis of the resonance fluorescence experiments have been developed in previous papers[4,5] for a single level decaying only by gamma-ray emission directly to the ground state. If, as in the case for As^{75}, gamma-ray branching occurs and if the internal conversion is not negligible, the expressions are slightly modified.[6] For convenience, the pertinent relations are repeated here for the case where the natural width Γ is small compared with the Doppler width $\Delta=(E_\gamma/c)(2kT/M)^{\frac{1}{2}}$ of the thermal agitation. Γ shall denote the total width of the level, i.e., $\Gamma=\sum(\Gamma_{\gamma\nu}+\Gamma_{\alpha\nu})$ where $\Gamma_{\gamma\nu}$ is the partial width of the νth gamma-ray transition and where $\Gamma_{\alpha\nu}$ is the partial width due to internal conversion of the νth transition. $\Gamma_{\gamma0}$ refers to the direct transition to the ground state.

The effective cross section for resonance scattering has the pure Doppler form

$$\sigma_D(E)=(g_2\lambda^2\Gamma_{\gamma0}{}^2/4g_1\pi^{\frac{1}{2}}\Gamma\Delta)\exp\{-[(E-E_r)/\Delta]^2\}. \quad (1)$$

Here g_2 and g_1 are the statistical weights of excited state and ground state, respectively, λ is the wavelength of the gamma radiation, E_r is the resonance energy, and E is the actual energy.

For a thick scatterer, the number of gamma rays scattered at a depth X out of an element dX by resonance fluorescence is

$$S(X)dX=ndX\int_0^\infty \sigma_D(E)N(E)\eta_e(E,X)\times\exp[-nX\sigma_D'(E)]dE, \quad (2)$$

where $N(E)$ is the number of incident gamma rays (at $X=0$) in the interval dE, n the number of nuclei per cm³ of the isotope under study, $\eta_e=\exp(-\sum n_i\sigma_i X)$

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† Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] A. W. Schardt and J. P. Welker, Phys. Rev. **99**, 810 (1955).

[2] A. W. Schardt, Phys. Rev. **108**, 398 (1957).

[3] See, e.g., Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. **28**, 432 (1956).

[4] F. R. Metzger, Phys. Rev. **101**, 286 (1956); and **103**, 983 (1956).

[5] C. P. Swann and F. R. Metzger, Phys. Rev. **108**, 982 (1957).

[6] J. D. Jackson, Can. J. Phys. **33**, 575 (1955).

the electronic attenuation, and $\sigma_D' = \sigma_D(\Gamma/\Gamma_{\gamma 0})$ the effective cross section for resonance absorption of the radiation of energy E. By expanding the exponential in (2) and making use of the slow variation with E of $N(E)$ and $\eta_e(E,X)$, the integration over E can be carried out with the result

$$S(X)dX = N(E_r)\eta_e(E_r,X)n\left(\frac{g_2\lambda^2\Gamma_{\gamma 0}{}^2}{4g_1\Gamma}\right) \times\left(1+\sum_{m=1}^{\infty}\frac{(-1)^m(nK'X)^m}{m!(m+1)^{\frac{1}{2}}}\right)dX, \quad (3)$$

where

$$K' = (g_2\lambda^2\Gamma_{\gamma 0}/4g_1\pi^{\frac{1}{2}}\Delta). \quad (4)$$

The series in the brackets represents the effect of selective absorption within the scatterer. The integration over the scatterer is usually done numerically.

For the self-absorption experiment, an absorber of thickness D is placed between the source and the scatterer. The contribution from the element dX of the scatterer is then obtained by replacing X in (3) by $X+D$.

If both a scattering experiment and a self-absorption experiment have been performed, it is usually advantageous to first analyze the self-absorption experiment. This determines K' which then can be used to take into account the self-absorption within the scatterer.

CHOICE OF COMPENSATION METHOD

In order to observe resonance fluorescence, when using radioactive isotopes as the sources of the exciting radiation, it is necessary to compensate for the recoil energy losses suffered in the emission and absorption processes.[7] This compensation could be realized by using the Doppler broadening form thermal agitation at high temperatures,[8] the Doppler shift produced by mechanical motion,[9] or the Doppler broadening arising in gaseous sources from the preceding radiation.[4] Of these methods, the last one appeared to be the most promising for the study of As^{75} using Se^{75} sources. The thermal method, although experimentally very simple, was rejected because, for the energies involved, it would give rise to rather small effects. Moreover, at the temperatures necessary for the thermal method (1000°C), all commonly available selenium compounds are in the gaseous phase. This means that, even with a low specific activity source, the recoils from the preceding radiation would alter the Maxwellian line shape and would render the interpretation difficult. The mechanical method was not considered because, even for the 265-kev transition, the peripheral speed of the centrifuge would have to be 1.2×10^5 cm/sec, i.e., very close to the limit set by the tensile strength of commonly available materials.

In the case of Se^{75} the conditions for the method utilizing the recoil from the preceding radiation in a gaseous source are very favorable. The energy of the neutrino accompanying the K-capture decay of Se^{75} is 455 kev, i.e., it is larger than the energy of the most energetic gamma ray present in the disintegration of Se^{75}. Therefore, for selenium compounds of low molecular weight ($M<90$), one will have an appreciable overlap of the absorption lines and the incident spectrum, and one can expect to observe resonance fluorescence from all those levels of As^{75} which have strong transitions to the ground state.

Elemental selenium is probably the simplest source to prepare, but the interpretation of the data is complicated by the presence of Se_6 and Se_2 molecules and of Se atoms in strongly temperature-dependent equilibria. For the study of the 265-kev transition, SeO_2 is another convenient source. Both the elemental Se source and the SeO_2 sources have to be kept at elevated temperatures in order to have all the activity in the gaseous phase.

For most of the experiments reported here, the radioactive selenium was used in the form of hydrogen selenide gas. It combines the convenience of being gaseous at room temperature with the low molecular weight of 77. It represents the closest approximation to monatomic selenium vapor which could be obtained with the available specific activity.

SOURCE PREPARATION

Selenium, enriched in Se^{74} (14.1%),[10] was irradiated for one month in the Materials Testing Reactor at Arco, Idaho. The specific activity of Se^{75} obtained in this way was twenty millicuries per milligram of enriched selenium.

For the preparation of the H_2Se sources, the selenium was distilled into a quartz ampoule, hydrogen gas in excess was added and the ampoule sealed off. The elements were then combined into H_2Se by heating the ampoule to 550°C. The gas decomposed at room temperature at a rate of less than one percent per day. Short heating to 550°C recombined the decay products again to H_2Se.

Sources with Se contents varying from 0.03 mg/ml to 0.5 mg/ml were prepared. The initial hydrogen pressure was chosen so as to leave approximately one tenth of atmospheric pressure of H_2 after formation of the H_2Se.

SCATTERING EXPERIMENT

A survey of the resonance fluorescence from arsenic was carried out in the usual ring geometry[4] with a

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[7] See, e.g., E. Pollard and D. E. Alburger, Phys. Rev. **74**, 926 (1948).

[8] K. G. Malmfors, Arkiv Fysik **6**, 49 (1952); F. R. Metzger and W. B. Todd, Phys. Rev. **95**, 853 (1954); F. R. Metzger, Phys. Rev. **97**, 1258 (1955) and J. Franklin Inst. 261, 219 (1956).

[9] P. B. Moon, Proc. Phys. Soc. (London) **A64**, 76 (1951); P. B. Moon and A. Storruste, Proc. Phys. Soc. (London) **A66**, 585 (1953); W. G. Davey and P. B. Moon, Proc. Phys. Soc. (London) **A66**, 956 (1953).

[10] Obtained from the Stable Isotopes Division of the U. S. Atomic Energy Commission, Oak Ridge, Tennessee.

source of H_2Se. A source of solid, elemental Se^{75} of equal strength was used for comparison purposes. Above 200-kev gamma-ray energy, the only sizable resonance effect was that from the 265- and 280-kev levels. Figure 1 shows the pulse-height distributions of the scattered radiation for the two selenium sources in the region of the (265+280)-kev full energy peak. The difference curve is compared in Fig. 2 with the pulse-height distribution obtained by exposing the scintillation counter to the direct gamma radiation from a source of Se^{75}. Contributions from the 402-, 305-, and 199-kev gamma rays were subtracted from the direct beam curve before it was normalized to the same area as the pulse-height distribution of the resonance radiation. The direct beam distribution is considerably wider than that expected for a single gamma ray. Using the 279-kev transition in Tl^{203} as a standard, the expected line shapes for the 265- and 280-kev transitions were obtained and the composite peak was analyzed into the contributions from the 265- and 280-kev transitions. With $\frac{1}{32}$ in. of cadmium and $\frac{1}{32}$ in. of lead surrounding the detector, the peak height of the 280-kev line amounted to 36% of the peak height of the 265-kev transition. This is in reasonable agreement with the 45.7% reported by Schardt and Welker[1] for the intensity of the 280-kev gamma ray relative to the 265-kev transition.

When the pulse-height distribution of the resonance scattered radiation was analyzed in the same manner, the peak height of the 280-kev line was found to amount only to $(2\pm2)\%$ of the peak height of the 265-kev line. This means that the 280-kev radiation is at least nine times less efficient in exciting resonance fluorescence than is the 265-kev transition. Considering the spins of the two levels and the fact that some of the 280-kev transitions are unable to cause resonance scattering because they are preceded by the 17 millisecond 305-kev state, the 265- and 280-kev transitions in As^{75} should, for a given lifetime, be approximately equally efficient in producing resonance fluorescence. Thus the small relative intensity of the 280-kev line in the resonance scattered radiation has to be attributed to a lifetime at least nine times longer than that of the 265-kev transition.

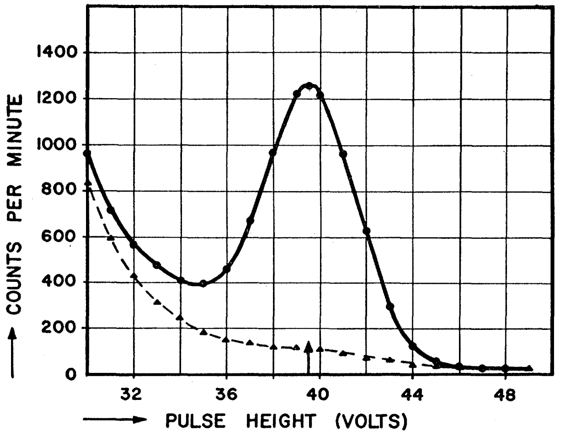

FIG. 1. Pulse-height distributions of Se^{75} radiation scattered from arsenic. Full circles were measured with H_2Se source, triangles with solid selenium source of equal strength. Arrow indicates expected position of 265-kev full energy peak.

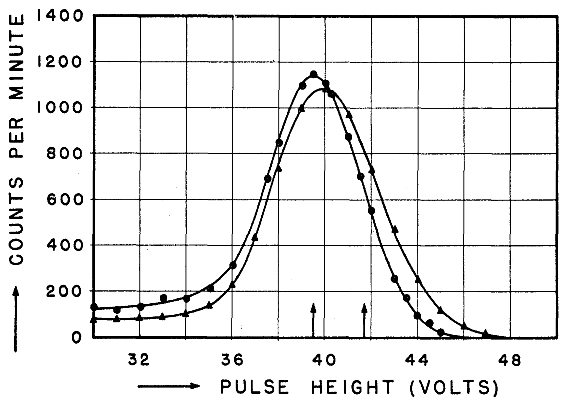

FIG. 2. Comparison of the pulse-height distributions of the resonance radiation and of the direct radiation from a Se^{75} source. Full circles represent the difference of the two curves of Fig. 1, i.e., the resonance radiation. Triangles refer to the direct beam after subtraction of the contributions from the 199-, 305-, and 402-kev transitions. Arrows indicate the expected positions of the 265- and 280-kev full energy peaks.

A study of the contribution from the 199-kev level was not attempted in view of the low intensity of the 199-kev line and because Compton scattering and resonance scattering from the other lines gave rise to large counting rates in the pulse-height region corresponding to the 199-kev full energy peak. Moreover, the excitation of the 265-kev level was expected to produce some 199-kev transitions via the 66-kev transition connecting the 265- and the 199-kev states.

Increasing the density of H_2Se in the source ampoules resulted in a reduction of the resonance fluorescence effect from the 265-kev level. For the lifetime of the 265-kev transition and the densities used, such a collision effect should not be noticeable. If, however, a much longer lifetime is postulated for the 402-kev level, then collisions will reduce the resonance effect because the 137-kev transition following the 402-kev level does not provide sufficient momentum to compensate for the recoil energy losses.

It is difficult to arrive at a reliable estimate of the lifetime of the 402-kev level on the basis of collision effects because of the complexity of the processes involved. A rough estimate placed the mean life of the 402-kev level between 3×10^{-10} and 3×10^{-9} second.

An attempt was made to measure the lifetime of the 402-kev level with the resonance fluorescence method. This method is not very sensitive for this particular level because the 402-kev ground-state transition, which can easily be detected, accounts only for 14% of the de-excitations. According to Eq. (3) the resonance scattering is proportional to $\Gamma_{\nu 0}^2/\Gamma$.

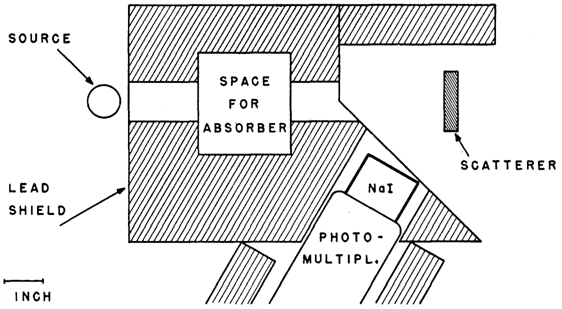

FIG. 3. Experimental arrangement used for the self-absorption experiment. Shielding on top of apparatus is omitted.

With $\Gamma_{402} \sim \Gamma/7$, the effect is about fifty times smaller than it would be if the level, with the same total width, deexcited exclusively to the ground state.

With a strong source of H_2Se ($\sim$20 millicuries) the counting rate in the 402-kev full energy peak did not exceed the background counting rate by a significant amount. From this, a lower limit of 10^{-10} second could be set for the mean life of the level, and a lower limit of 7×10^{-10} second for the partial lifetime of the 402-kev transition. Recently,[11] Day has shown that the half-life of the level is shorter than 2×10^{-9} second. It is thus possible to bracket the lifetime of the 402-kev state between 10^{-10} and 3×10^{-9} second.

Preliminary experiments[12] had indicated an approximately isotropic distribution of the 265-kev resonance radiation. However, the design of the scatterer used for these studies gave rise, for the low-energy gamma rays of Se^{75}, to appreciable attenuation of the incoming and outgoing gamma rays in the material containing the arsenic. It was, therefore, considered probable that the first angular distribution data had been somewhat distorted. Consequently, a new scatterer was built and the angular distribution measurements were repeated. The arsenic of the new scatterer was contained in 1/64 in. aluminum on all sides, the different parts being butt-jointed with Araldite cement. The experiments performed with this improved scatterer gave the following angular distribution of the 265-kev resonance radiation: $W(\theta) = 1 + (0.15 \pm 0.02) P_2(\cos\theta)$. The probable error of 0.02, estimated on the basis of the consistency of data taken with different sources, is considerably larger than the statistical uncertainty. The main difficulty seemed to arise from differences in the distribution of the activity in the gaseous sources and in the solid Se^{75} comparison sources.

SELF-ABSORPTION EXPERIMENTS

The magnitude of the resonance fluorescence from the 265-kev level made it possible to forego the use of the ring geometry for the self-absorption studies and to use the arrangement shown in Fig. 3. The scatterer was a cylinder of arsenic, 1.5 in. in diameter and 5/16 in. long, contained in 7-mil aluminum. Absorbers of up to 16 g/cm² of arsenic could be inserted between source and scatterer. Zinc was used for the comparison absorbers which were matched with the arsenic absorbers as far as the electronic absorption was concerned.

The absorption curve is shown in Fig. 4. The best fit to the experimental points, using Eq. (3), was obtained with $K' = 5.25 \times 10^{-24}$ cm². Upon assuming spin $\frac{3}{2}$ for the 265-kev level and using an effective temperature of 318° K, a mean life, τ_γ, of 1.64×10^{-11} second was calculated. The probable error is estimated to be $\pm 0.2 \times 10^{-11}$ second. Because of the low Debye temperature of arsenic ($\theta = 285°$)[13] the effective temperature, calculated according to Lamb,[14] is not much different from the actual temperature (300° K). The uncertainty introduced through the Doppler width Δ should, therefore, be rather small.

At the time these self-absorption experiments were carried out, it was felt that it might be possible to test Lambs correction in the case of arsenic. For this purpose, further self-absorption experiments were carried out with both the absorbers and the scatterer being held at liquid nitrogen temperature. The same apparatus (Fig. 3) was used, the scatterer and absorbers being contained in Styrofoam[15] vessels which could be filled with liquid nitrogen. Because of the space required for these containers, the absorber thickness had to be restricted to 8 g/cm².

On the basis of the actual temperatures—room temperature and liquid nitrogen temperature—a ratio of 1.93 was expected for the corresponding widths. With the effective temperatures calculated according

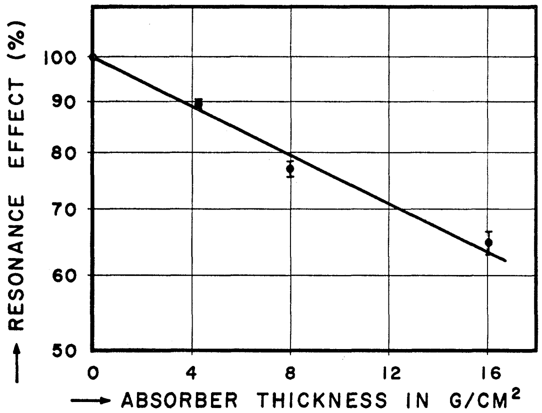

FIG. 4. Attenuation of the resonance scattering from 1.63-gram/cm² arsenic scatterer by arsenic absorbers inserted between source and scatterer. The almost straight line represents the best fit to the experimental points using Eq. (3).

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[11] R. B. Day (private communication).

[12] F. R. Metzger, J. Franklin Inst. **262**, 229 (1956).

[13] J. de Launay, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2, p. 233.

[14] W. E. Lamb, Phys. Rev. **55**, 190 (1939).

[15] Trade mark of the Dow Chemical Company.

to Lamb the expected ratio was 1.59. The experimental value, 1.71±0.20, seems to favor the value predicted by Lamb, but the large uncertainty prevents one from drawing any further conclusions.

RESULTS AND DISCUSSION

The results of the resonance fluorescence experiments, as far as the lifetimes of the transitions in As^{75} are concerned, are summarized in Table I.

Some of the consequences of the lifetime limits on the spin and parity assignments have already been discussed by Schardt.[2] The value for the lifetime of the 265-kev transition is that obtained from the self-absorption studies assuming spin $\frac{3}{2}$. The scattering experiments with the low-density sources led to a mean life of 2.5×10^{-11} second for the 265-kev transition, using a value $\Gamma_{\gamma 0}/\Gamma = 0.96$. The discrepancy between the two values of τ_γ indicates that collisions in the 402-kev state are effective in reducing $N(E_r)$ even for the low-density sources.

In arriving at the lifetime limits of Table I, the reduction of the resonance effects by collisions was assumed to be the same for all transitions.

From a comparison of the actual mean life of the 265-kev transition with the lower limit for the partial $E2$ lifetime (10^{-9} second) it is evident that the 265-kev transition is almost pure $M1$, i.e., $E2/M1 < 0.02$. This is in accord with the angular distribution data which, independant of whether the spin of the 265-kev level is $\frac{3}{2}$ or $\frac{5}{2}$, call for an intensity ratio $E2/M1 < 0.001$. The reported conversion coefficient[1] is considerably larger than the theoretical coefficient for a pure $M1$. If the theoretical $M1$ coefficient is accepted for the 265-kev transition, and preliminary conversion measurements with a lens spectrometer justify this, the experimental conversion coefficient[1] for the 280-kev transition can be corrected using the correction factor from the 265-kev transition. The corrected conversion coefficient of the 280-kev gamma-ray transition corresponds to an intensity mixing ratio $E2/M1 \approx 0.2$. Such a ratio can be accommodated within the upper and lower limits established for the lifetime of the 280-kev level without contradicting the partial $E2$ lifetime established in the Coulomb excitation experiments.[3] The ratio would also fit the (121, 280) angular correlation data[1] for a spin sequence $\frac{5}{2}$-$\frac{5}{2}$-$\frac{3}{2}$ using an almost pure 121-kev $E1$ transition.

TABLE I. Information on the mean lives of transitions in As^{75} obtained from the resonance fluorescence studies.

E_γ (kev)	τ_γ (sec)
265	$(1.6\pm 0.2)\times 10^{-11}$
280	$\geq 10^{-10}$
402	$\geq 7\times 10^{-10}$

The small quadrupole to dipole ratio suggested by the angular distribution measurements might tempt one to assume that the 265-kev transition is $E1$. However, this possibility seems to be ruled out by the small ft-value of the beta-ray spectrum feeding the 265-kev level in the decay of Ge^{75}.

If the reported conversion coefficient[1] of the 136-kev transition is also assumed to be too large, the angular correlation[1] of the 136- and 265-kev gamma rays is no longer in disagreement[2] with the small $E2/M1$ ratio of the 265-kev transition expected on the basis of the resonance fluorescence experiments.

From a survey of the information available it appears that accurate conversion data are at present the experimental evidence which is most necessary for a reliable classification of the levels of As^{75} and which is still not available.

ACKNOWLEDGMENTS

The author wishes to thank Mr. W. B. Todd for his assistance in all phases of the experimental work.

It is a pleasure to acknowledge helpful discussions with Dr. Alois W. Schardt.