Mean Life of the 265-keV Excited State in As^{75}†

FRANZ R. METZGER
Bartol Research Foundation of the Franklin Institute, Swarthmore, Pennsylvania

(Received January 12, 1962)

Using an ultracentrifuge as the means of imparting velocities of up to 11.5×10^4 cm/sec to a Se^{75} source, a resonance transmission experiment involving the 265-keV excited state in As^{75} has been carried out. If a branching ratio Γ_0/Γ of 0.96 is assumed for this state, the transmission vs velocity data lead to a mean life $\tau_{\text{level}}=(1.57\pm0.10)\times10^{-11}$ sec. This value agrees with the result of an earlier self-absorption study, but differs considerably from the mean lives deduced from resonance scattering experiments.

I. INTRODUCTION

IN resonance scattering experiments with gamma rays, the resonant process competes with the rather small nonresonant elastic scattering processes as, for instance, Rayleigh and Thomson scattering. In transmission experiments, on the other hand, the resonant process competes with the total electronic cross section which is usually several orders of magnitude larger than the nonresonant elastic cross section. In general, the scattering experiment is, therefore, a much more sensitive means of observing resonance fluorescence than is the transmission study. The main advantages of the transmission experiment are its simplicity and modest source strength requirements as well as the fact that all de-excitations, including internal conversion, are effective at the same time, while the scattering experiment concerns itself usually only with one mode, the gamma transition to the ground state.

For the resonance attenuation to compete successfully with the electronic attenuation, it is necessary to concentrate the integrated resonance cross section in as narrow an energy interval as possible. The most favorable example is provided by the recoil-free emission lines for which the transmission experiment is standard procedure. The next highest concentration of gamma rays per unit energy interval is found in the free-recoil emission lines having the pure "Doppler form"

$$N(E)=(N_0/\Delta\pi^{\frac{1}{2}})\exp\{-[(E-E_c)/\Delta]^2\}, \quad (1)$$

where $\Delta=E_\gamma(2kT/Mc^2)^{\frac{1}{2}}$ is called the Doppler width, and where E_c is the energy at the center of the line. The centrifuge method, i.e., the method employing the high speeds attained with rotors for the purpose of compensating for the recoil energy losses, utilizes such Doppler-broadened lines and makes E_c equal to the resonance energy E_r by the proper choice of the source (rotor) velocity. One would, therefore, expect the transmission method to have been widely used in studies using the centrifuge compensation technique. However, so far, not a single transmission experiment of this type had been performed. The reason for this may be traced to the restrictions with respect to transition energies and atomic weights imposed on the centrifuge method by the tensile strength to density ratios of commonly available rotor materials. For presently available rotors, the gamma energies that can be studied are limited by the condition

$$E_\gamma\ (\text{keV})\lesssim 3.7\times A,$$

where A is the atomic weight of the nucleus in which the transition takes place. This condition, and the trend in level spacings with A, strongly favor heavy nuclei as candidates for the centrifuge method, i.e., they favor those nuclei for which the transmission experiment is difficult because of the large photoelectric cross sections. Even if one managed to stay away from the heavy nuclei, one would be faced with the fact that the restriction to low-energy transitions is also a restriction to small natural widths and thus to small resonance cross sections. The importance of this fact is illustrated by the success of transmission experiments with high-energy transitions from (p,γ) reactions for which the large natural widths offset the disadvantages of emission lines which are considerably broader than the pure thermal Doppler lines.

The 265-keV transition in As^{75} is a very favorable case for a transmission experiment for three reasons: The high abundance (100%) of As^{75}, the low atomic number (33) of arsenic, and the relatively short lifetime. This lifetime had been determined by resonance scattering and by self-absorption experiments. Two independent scattering experiments[1,2] using gaseous H_2Se^{75} sources led to mean lives of 2.5×10^{-11} and $(2.3\pm0.3)\times10^{-11}$ sec. A self-absorption study[1] yielded a mean life $\tau_\gamma=(1.6\pm2.0)\times10^{-11}$ sec. The difference between these two sets of values, though small in the light of present day uncertainties in the theoretical predictions, was considered sufficiently large to warrant another determination with a third resonance fluorescence method. For this purpose, the centrifuge method offered the advantage of being able to use a solid source for which the shape of the emission line was known and could also be verified experimentally. The transmission technique added the advantage of not requiring any knowledge of the angular distribution of the resonance radiation.

In the following sections, a description of the experi-

† Supported by a grant from the National Science Foundation.

[1] F. R. Metzger, Phys. Rev. **110**, 123 (1958).
[2] H. Langevin-Joliot, M. Langevin, J. phys. radium **19**, 765 (1958).

mental arrangement will be given; the data obtained with As^{75} will then be presented; the procedure used in the analysis of the data will be discussed in some detail; and finally, an attempt will be made to discuss the consequences of the disagreement with some of the previous studies, and to judge the general usefulness of the transmission method as applied to the centrifuge technique.

II. EXPERIMENTAL PROCEDURES

Figure 1 shows the general arrangement of source, collimator, and detector. The star-shaped rotors, machined from titanium alloy C-135AMo [3] were coupled to an air-driven 0.5-in.-diam turbine by means of a 0.071-in.-diam piano wire shaft. The vacuum seal was made in a Babbitt metal[4] bearing by DuoSeal pump oil which served at the same time as a lubricant. The rotor speed was controlled by regulating the air flow to the turbine. With standard commercial regulators the speed could easily be kept constant to better than 1%. The detector viewed the source through a lead collimator, which to some extent defined the source path over which the absorption was averaged. The actual definition was achieved by gating the electronics, the gate originating from two photomultiplier tubes which received light signals from a mirror attached to the drive shaft. An absorber with a surface density of 16.05 g/cm^2 of arsenic and a zinc absorber were alternately inserted into the beam for counting periods of 2 and 4 min. Pulses falling within $\pm 5\%$ of the 265-keV full-energy pulse height were accepted when the data at the different source velocities were taken. At the top speed of 11.46×10^4 cm/sec, another series of runs at different pulse heights was carried out with a narrower channel ($\pm 2\%$). This series served as a check of the reliability of the decomposition of the (265+280)-keV peak into its components. In carrying out this decomposition, the pulse-height distribution for the single line from a Hg^{203} source was most useful, since the energy of this transition in Tl^{203} agrees very closely with the energy of the 280-keV transition in As^{75}.

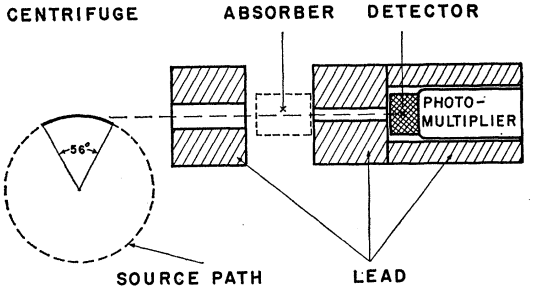

FIG. 1. Experimental arrangement for the transmission experiment. Counts were accepted only while the source moved along the heavily drawn 56-deg portion of its 12.1-cm diam path.

[3] Manufactured by the Crucible Steel Company of America.
[4] Magnolia antifriction metal.

With the wide channel ($\pm 5\%$), a comparison of the counting rates with the arsenic and the zinc absorbers was also carried out at zero speed, i.e., with the source at rest at several positions of its path. Since the resonance effect for zero speed is negligible, the experimental difference of (2.9 ± 0.3) percent in the counting rates was attributed to an actual mismatch of the two absorbers with respect to nonresonant absorption. The data subsequently collected at fifteen different source speeds were corrected accordingly.

III. EXPERIMENTAL RESULTS

From the decomposition of the pulse-height distribution it was concluded that, with the absorber in the beam, but in the absence of resonant absorption, the 265-keV transition contributed (71 ± 4) percent of the counting rate in the wide ($\pm 5\%$) channel. This value of 71% was used in arriving at the results of the absorption vs velocity measurements summarized in Table I. The errors quoted in the table are the statistical errors only, the uncertainty in the fraction of 265-keV counts was not included.

The series of measurements carried out at the source velocity of 11.46×10^4 cm/sec with a narrow ($\pm 2\%$) detection channel gave an absorption of (35.8 ± 1.4) percent, in good agreement with the value (36.2 ± 1.9) percent obtained with the wide channel.

IV. EVALUATION

In analyzing experimental results obtained with the centrifuge method, it is customary to use what is sometimes called the effective cross section for resonance scattering, i.e., the cross section averaged over the

TABLE I. Experimental results of the transmission experiments carried out with the 16.05-g/cm^2 arsenic absorber. The absorptions expected for $\Gamma_0=4.07\times10^{-5}$ eV are tabulated for comparison purposes. In the last column, the widths calculated from the observed absorptions are listed.

Source velocity (10^4 cm/sec)	Absorption in % Experiment	Absorption in % Calculated for $\Gamma_0=4.07\times10^{-5}$ eV	Width Γ_0 from experimental absorption (10^{-5} eV)
1.89	-0.9 ± 2.0	0.07	-55 ± 120
2.84	0.7 ± 1.4	0.24	12 ± 65
3.79	0.9 ± 2.0	0.7	5.4 ± 12
4.74	0.4 ± 1.4	1.7	1.0 ± 3.4
5.69	4.4 ± 2.1	3.7	4.8 ± 2.4
6.65	4.1 ± 1.6	7.4	2.1 ± 0.9
7.60	14.1 ± 1.9	12.6	4.7 ± 0.8
8.08	17.4 ± 2.1	16.0	4.5 ± 0.7
8.56	18.6 ± 2.1	19.6	3.8 ± 0.6
9.04	23.0 ± 2.1	23.4	4.0 ± 0.4
9.53	23.3 ± 2.1	27.2	3.3 ± 0.4
10.01	33.0 ± 1.9	30.5	4.5 ± 0.3
10.49	32.7 ± 1.9	33.4	4.0 ± 0.3
10.97	37.8 ± 1.9	35.6	4.4 ± 0.3
11.46	36.2 ± 1.9	36.8	4.0 ± 0.3
11.93	...	37.3	...
12.41	...	36.5	...
13.37	...	32.4	...

incident line. For the case of pure resonance fluorescence, i.e., if the gamma transition to the ground state is the only mode of decay ($\Gamma_0=\Gamma$), this cross section is given by

$$\sigma_{av}=\frac{g_2}{g_1}\frac{\lambda^2\Gamma}{4\pi^{\frac{1}{2}}E_\gamma}\left[\frac{Mc^2}{2k(T_1+T_2)}\right]^{\frac{1}{2}} \times\exp\left[-\frac{M(E_\gamma/Mc-u)^2}{2k(T_1+T_2)}\right], \quad (2)$$

where g_2 and g_1 are the statistical weights of excited state and ground state, respectively, λ is the wavelength of the radiation of energy E_γ, Γ is the total width of the level; M the mass of the recoiling nucleus, k Boltzmann's constant, c the velocity of light, T_1 and T_2 the absolute effective temperatures of source and absorber (scatterer), and u is the speed of approach of source to absorber (scatterer).

The use of Eq. (2) is justified as long as the resonance attenuation in the absorber (scatterer) is small, i.e., as long as the line shape of the incident radiation is not appreciably modified in the course of its passage through the absorber (scatterer). For the transitions which had been studied in the past,[5] this condition was fulfilled because the transitions were either slow and/or occurred in isotopes of small abundance, and because heavy nuclei ($Z\geq 59$) were involved. For the large absorptions observed with As^{75}, Eq. (2) may be used only for preliminary estimates; for the final evaluation, the shapes of the absorption line and of the incident spectra have to be taken into account in more detail. Figure 2, which is based on such a detailed calculation, shows the degree of modification of the Se^{75}-As^{75} gamma radiation through resonance attenuation in 16.05-g/cm² of arsenic.

For the more detailed calculations in the general case where $\Gamma_0\neq\Gamma$, one may start out with the resonance absorption cross section[6] for photons of energy E:

$$\sigma_{abs}(E)=(g_2/g_1)(\lambda^2/8\pi)\Gamma_0\Gamma/[(E-E_r)^2+\tfrac{1}{4}\Gamma^2], \quad (3)$$

where Γ_0 is the partial width for gamma de-excitation to the ground state, Γ is the total width of the level, and E_r the resonance energy. The integrated cross section is then

$$\sigma_{abs}(E)dE=g\lambda^2\Gamma_0/4, \quad (4)$$

where g stands for the ratio g_2/g_1 of the statistical weights. Note that this cross section does not depend on the total level width Γ.

In the free-recoil case with which we are concerned here, the thermal motion of the absorbing nuclei has the effect of spreading the resonance energies in the laboratory system over a range of energies which is

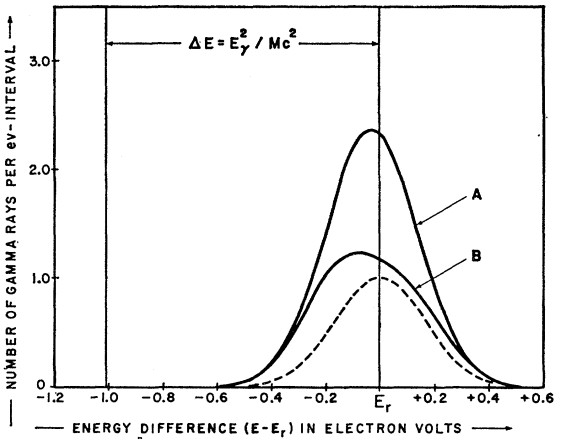

FIG. 2. Effect of the resonance attenuation in 16.05-g/cm² of As on the energy distribution in the gamma line. Curve A represents the incident line for $u=11.46\times10^4$ cm/sec, curve B the transmitted line if $\Gamma_0=4.07\times10^{-5}$ eV. The absorption line is indicated by the dashed line.

usually several orders of magnitude larger than the natural width. The shape of the absorption line for an ensemble of nuclei is then determined by the thermal velocity distribution, i.e., it has the "Doppler form":

$$\sigma_D(E)=\frac{\int\sigma(E)dE}{\Delta\pi^{\frac{1}{2}}}\exp\{-[(E-E_r)/\Delta]^2\} =g\frac{\lambda^2\Gamma_0}{4\Delta\pi^{\frac{1}{2}}}\exp\{-[(E-E_r)/\Delta]^2\}. \quad (5)$$

According to Lamb,[7] the effect of the crystalline binding on the width of the thermal distribution may be taken into account by using for T not the actual temperature, but an effective temperature related to the actual temperature via the Debye temperature of the crystalline material. In the As^{75} experiment, the actual absorber temperature was 302°K. With a Debye temperature $\theta=285°$,[8] one finds on the basis of Lamb's treatment an effective temperature $T=316$°K. With this value, the Doppler width of the 265-keV line in As^{75} becomes $\Delta=0.234$ eV.

The incident 265-keV radiation from a solid source of Se^{75} moving towards the absorber (scatterer) with a velocity u has the energy distribution

$$N(E)=(N_0/\Delta)\pi^{-\frac{1}{2}}\exp\{-[(E-E(u))/\Delta]^2\}. \quad (6)$$

Here, N_0 is the total number of gamma rays, and Δ is the Doppler width for the source material. Although the Debye temperature of Se is lower than that of As, the difference was just about canceled by the slightly higher temperature of the source ($\approx$312°K). Consequently, the same effective temperature (316°K) was

[5] See Table II.

[6] See, for instance, F. R. Metzger, *Progress in Nuclear Physics*, edited by O. R. Frisch (Pergamon Press, New York, 1959), Vol. 7.

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

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

used for the source as well as for the absorber. The line center of the incident line, $E(u)$, is related to the energy E_r of the center of the absorption line, in the approximation used here, by

$$E(u)=E_r(1-E_r/Mc^2+u/c). \quad (7)$$

Since the transmission was measured for source positions ranging from $-28°$ to $+28°$ relative to the tangential position, the effective source velocity components ranged from $0.883\times u$ to u. Because of this spread in the velocities, the actual energy distribution of the 265-keV radiation differed slightly from that given by Eq. (6). For three velocities, $u=7.6\times10^4$, 9.53×10^4, and 11.46×10^4 cm/sec, the shape of the incident energy distribution $N(E)$ was calculated exactly. It was found that the line shape obtained in this way could be reproduced with sufficient accuracy by Eq. (6), if u was replaced by $0.96\times u$ and Δ by $1.02\times\Delta$. This procedure was then adopted for the other velocities.

The shapes $N(E)$ were then combined with the Doppler cross section of Eq. (5) to calculate the transmitted lines and obtain the absorptions

$$\text{Abs}=(1/N_0)\int N(E)\{1-\exp[-nt\sigma(E)]\}dE, \quad (8)$$

for an absorber with nt nuclei per cm^2. These calculations were carried out for different values of the source velocity, and at each velocity for several values of the width Γ_0. This enabled one to plot, for each velocity u, the absorption vs Γ_0. Each experimental absorption could then be assigned a value of Γ_0; these values of Γ_0 are listed in the last column of Table I. The mean value of Γ_0 obtained from that column was $\Gamma_0=(4.07\pm0.13)\times10^{-5}$ eV. In Fig. 3, the absorptions expected for this value of Γ_0 are compared with the experimental points. The agreement in the shape of the absorption vs source velocity is very satisfactory. This fact is also indicated by the absence of any significant trend with source velocity in the widths listed in the last column of Table I.

From the series of runs carried out at 11.46×10^4 cm/sec with the narrow ($\pm2\%$) channel, a width $\Gamma_0=(3.94\pm0.21)\times10^{-5}$ eV was obtained. If this value is combined with the result of the wide ($\pm5\%$) channel measurements, a grand mean,

$$\Gamma_0=(4.03\pm0.11)\times10^{-5}\text{ eV},$$

is obtained. Using $\Gamma_0/\Gamma=0.96$, one calculates from this Γ_0 a total width $\Gamma=(4.20\pm0.12)\times10^{-5}$ eV for the 265-keV level in As^{75}. The corresponding mean life of the level is $\tau=(1.57\pm0.05)\times10^{-11}$ sec. If the statistical uncertainty is combined with the error in the 265/280 intensity ratio, the final value for the mean life of the level becomes

$$\tau_{\text{level}}=(1.57\pm0.10)\times10^{-11}\text{ sec}.$$

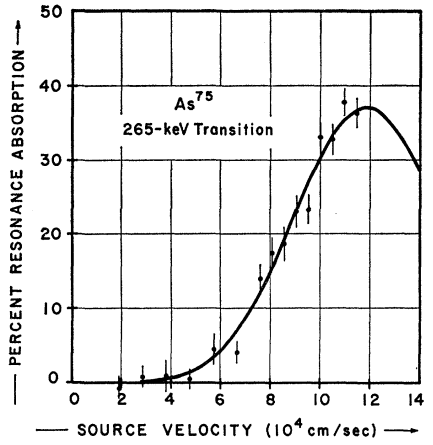

FIG. 3. Comparison of the experimental absorption points (with their statistical uncertainties) with the absorption calculated for $\Gamma_0=4.07\times10^{-5}$ eV.

V. DISCUSSION

The new value for the mean life of the 265-keV level in As^{75}, $\tau=(1.57\pm0.10)\times10^{-11}$ sec, agrees very well with the result of the previous self absorption study,[1] $\tau=(1.57\pm0.20)\times10^{-11}$ sec, but disagrees with the average of the two simple scattering studies,[1,2] $\tau=(2.3\pm0.2)\times10^{-11}$ sec. While this result is gratifying as far as the reliability of self-absorption studies is concerned, it emphasizes anew the difficulties one often faces in the evaluation of simple scattering information for sources utilizing recoils from preceding radiation. In fairness to the simple scattering experiment, it should be mentioned that the situation in As^{75} is especially unfavorable because of the existence of the long lived 402-keV state through which most of the Se^{75} decays proceed to the 265-keV level. Since the recoil from the neutrino involved in the K capture leading to the 402-keV level is needed for the restoration of the resonance condition for the 265-keV radiation, collision effects during the mean life[2] $\tau=(1.5\pm0.4)\times10^{-9}$ sec of the 402-keV level have to be minimized through the use of low source pressures. At these low pressures, the fraction of the activity adhering to the walls of the source container might no longer be negligible. Because recoils going into the walls are slowed down completely before the emission of the 265-keV radiation and are essentially not contributing any resonant gamma rays, the activity on the walls provides a smaller fraction of resonant gamma rays than does the activity which is in the gaseous phase. With some activity adhering to the walls, extrapolation to zero pressure, i.e., to the collision-free case, of the fraction of resonant gamma rays in the entire source will, therefore, give an incorrect value for this fraction and with it for the mean life of the 265-keV level. It was for reasons like this that reference 1 quoted the result of the self-absorption study and did not give any weight to the result of the scattering experiment. The experience with As^{75} points

TABLE II. Expected resonance absorptions, for absorbers of one-half value thickness for electronic attenuation, for all the transitions studied so far with the centrifuge method.

Isotope	Abundance (%)	E_γ(keV)	Γ_0 (eV)	Resonance absorption (%)
As^{75}	100	265	4.0×10^{-5} [a]	17.1
Pr^{141}	100	145	2.3×10^{-7} [b]	0.07
Gd^{155}	15.1	105	1.1×10^{-6} [c]	0.05
Lu^{175}	97.4	343	1.4×10^{-6} [d]	0.07
Hg^{198}	10.0	411	2.1×10^{-5} [e]	0.45
Hg^{199}	16.8	208	3.9×10^{-6} [f]	0.11
Tl^{203}	29.5	279	1.3×10^{-6} [g]	0.05

[a] This paper.
[b] F. R. Metzger, J. Franklin Inst. **261**, 219 (1956).
[c] B. I. Deutch, F. R. Metzger, and F. J. Wilhelm, Nuclear Phys. **16**, 81 (1960).
[d] B. I. Deutch, Nuclear Phys. **30**, 191 (1962).
[e] W. G. Davey and P. B. Moon, Proc. Phys. Soc. (London) **A66**, 956, (1953).
[f] V. Knapp, Proc. Phys. Soc. (London) **A70**, 142 (1957).
[g] B. I. Deutch and F. R. Metzger, Phys. Rev. **122**, 848 (1961).

out the importance of carrying out self-absorption studies whenever they are feasible and an uncertainty concerning the shape of the incident line exists.

As far as the combination of transmission studies with the centrifuge technique is concerned, the As^{75} experiment confirmed the expectation that, for large absorptions, the transmission study is a very efficient way of measuring cross sections. All the data reported in this paper, for instance, was accumulated in less than 10 hours of centrifuge operation. Unfortunately, most absorption effects encountered in typical centrifuge experiments are rather small. This fact is illustrated in Table II, where all the isotopes studied so far with the centrifuge method are listed, and where the resonance absorption effects, expected for absorbers of one-half value thickness for electronic attenuation, are given. It becomes clear from this tabulation that As^{75} is indeed a very favorable case, and that transmission studies with any of the other isotopes would be, to say the least, very tedious.

As improved rotors become available, studies with lower Z nuclei will become more feasible, and the opportunity for transmission studies will grow. Higher rotor speeds are often achieved at a sacrifice of space available for the source. Since the activities required for transmission experiments are approximately one order of magnitude smaller than those necessary for scattering studies, the trend towards higher rotor speeds will favor once more the transmission-type experiment.

A transmission study is also indicated whenever Γ_0/Γ is small, i.e., for large branching (internal conversion), since the absorption is proportional to Γ_0 while the scattering is proportional to Γ_0^2/Γ.

PHYSICAL REVIEW VOLUME 127, NUMBER 1 JULY 1, 1962

Study of the $Al^{27}(\alpha,Be^7)Na^{24}$ Reaction from Threshold to 41 MeV*

NORBERT T. PORILE
Chemistry Department, Brookhaven National Laboratory, Upton, New York
(Received February 16, 1962)

Excitation functions for the formation of Na^{24}, Be^7, and Na^{22} in the interaction of Al^{27} with alpha particles have been measured from threshold to 41 MeV. The cross sections at 41.2 MeV are 0.33, 0.14, and 8.0 mb, respectively, and the excitation functions exhibit a sharp rise with bombarding energy. The recoil properties of the above nuclides have been investigated by means of thick-target integral range measurements at 40 MeV. The results for Be^7 are consistent with approximately equal contributions from direct interaction and evaporation processes while the results for Na^{24} and Na^{22} indicate that these nuclides are primarily formed through processes involving compound-nucleus formation. It is concluded that at 40 MeV approximately 60% of the yield of Na^{24} may be attributed to the $(\alpha,\alpha He^3)$ reaction.

I. INTRODUCTION

THE formation of Be^7 in medium-energy nuclear reactions was first studied by Bouchard and Fairhall.[1] These authors detected Be^7 as a product of the reactions of oxygen, aluminum, and copper with 33–42 MeV alpha particles. The formation of Be^7 in the bombardment of aluminum with alpha particles is of particular interest since Na^{24}, the complementary product in the reaction, may also be detected by radiochemical techniques. As a result, a detailed study of this reaction is possible. Bouchard and Fairhall[1] thus reported approximately equal cross sections for the formation of Be^7 and Na^{24} from Al with 40-MeV alpha particles. These workers also studied the emission of Be^7 recoils in the forward and backward directions and on the basis of the absence of backward emission concluded that the $Al^{27}(\alpha,Be^7)Na^{24}$ reaction proceeds by a pickup mechanism.

More recently, Lindsay and Carr[2] also investigated

* Research performed under the auspices of the U. S. Atomic Energy Commission.
[1] G. H. Bouchard, Jr. and A. W. Fairhall, Phys. Rev. **116**, 160 (1959).
[2] R. H. Lindsay and R. J. Carr, Phys. Rev. **120**, 2168 (1960).