Determination of the Lifetime of the 0.559-MeV Level of Se^{76} using Nuclear Resonance Fluorescence*

JOHN R. PRUETT
Bryn Mawr College, Bryn Mawr, Pennsylvania
(Received 23 October 1962)

The lifetime of the 0.559-MeV 2^+ level of Se^{76} has been measured using a nuclear resonance fluorescence technique. In this work a gaseous source of AsH_3 was employed and the 0.559-MeV gamma rays emitted following the beta decay of As^{76} were scattered from a Se scatterer and from a comparison Zn scatterer. From the difference in the observed scattering, the average resonant scattering cross section for the emitted gamma ray line was obtained. With an estimate of the ratios of the nuclear matrix elements for the As^{76} beta decay, the fraction $N(E_{res})/N$ of the 0.559-MeV gamma rays having energies that differ by 0.5 eV or less from the resonant energy is computed, and the lifetime of the 0.559-MeV state is calculated. The result obtained is $\tau_{0.559}=(1.55_{-0.19}^{+0.13})\times10^{-11}$ sec. In a similar way, an estimate of the lower limit of the lifetime of the 1.216-MeV level of Se^{76} is found to be $\tau_{1.216}>3\times10^{-12}$ sec.

I. INTRODUCTION

NUCLEAR resonance scattering and resonance absorption techniques have been used by numerous investigators to obtain information concerning lifetimes, spins, and magnetic moments of excited nuclear states. The results reported here are actually a by-product of more extensive investigations designed to test the feasibility of using resonance fluorescence techniques to obtain information about the recoil momentum distribution and nuclear spin polarization of nuclei recoiling from a previous beta-decay process. Such information could be used to help determine nuclear matrix elements in some forbidden beta-decay processes. The results of this investigation will be reported in a later paper.

In attempting any nuclear resonance fluorescence experiment it must be recognized that the energy lost through recoil in the emission and subsequent absorption process will prevent any resonance fluorescence through a given excited nuclear state by gamma rays emitted from the same nuclear state in a source, unless some method is used to re-establish resonance by compensating for this lost recoil energy.[1] Several methods have been used to establish the resonance condition all depending upon an increase in the energy of the emitted gamma ray at the expense of kinetic energy stored in the motion of the emitting nucleus prior to the emission process. If the component of momentum of the nucleus in the direction of gamma emission is equal to the momentum of the subsequently emitted gamma ray, the extra energy received by this gamma photon is just equal to that which will go into recoil of an absorbing nucleus if the gamma is "resonant captured." The necessary source velocity component has been obtained through the use of the thermal velocity of atoms in a solid or liquid source at high temperature,[2,3] by giving the entire source the necessary velocity as a result of attaching it to the rim of a spinning wheel or centrifuge disk,[4,5] or by using a gaseous or liquid source in which the recoil momentum resulting from a previous emission process will not be lost before the gamma ray is emitted.[6,7] The present work was carried out using recoil from a previous beta decay with the source in the form of arsine gas (AsH_3) at a pressure of a few millimeters of mercury. This same technique was employed by Metzger[7] and the experimental setup used in the following work is similar to his.

The line shape for gamma rays emitted from nuclei recoiling from a previous beta decay of around 2.5-MeV energy release will have a width of the order of twenty electron volts due to Doppler broadening from the recoil velocity, whereas the width of the absorption line when corrected for the Doppler broadening due to the thermal motion in the scatterer will be of the order of an electron volt. This means that only a small region of the emitted line will contribute to any absorption or scattering process. Thus to relate an experimentally determined average cross section $\bar{\sigma}$ to the level width of the nuclear state through which scattering is taking place will require knowledge of the fraction $N(E_{res})/N$ of all of the gamma rays in the emitted line which fall within an electron volt interval around the peak of the absorption line. When the thermal broadening of the absorption line in the scatterer is taken into account and when $\bar{\sigma}$ represents a cross section averaged over all atoms in a scatterer which has a mixture of isotopes, with A_d being the fractional abundance of those atoms from which resonance scattering can take place, the average resonant scattering cross section is given by

* This work was supported by a grant from the National Science Foundation.

[1] K. G. Malmfors, in *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 18(II), p. 521.

[2] K. G. Malmfors, Arkiv Fysik **6**, 49 (1952).

[3] Franz R. Metzger and W. B. Todd, Phys. Rev. **95**, 853 (1954).

[4] P. B. Moon, Proc. Phys. Soc. (London) **63**, 1189 (1950).

[5] B. I. Deutch and F. R. Metzger, Phys. Rev. **122**, 848 (1961).

[6] K. Ilakovac and P. B. Moon, Phys. Rev. **93**, 254 (1954).

[7] Franz R. Metzger, Phys. Rev. **101**, 286 (1956).

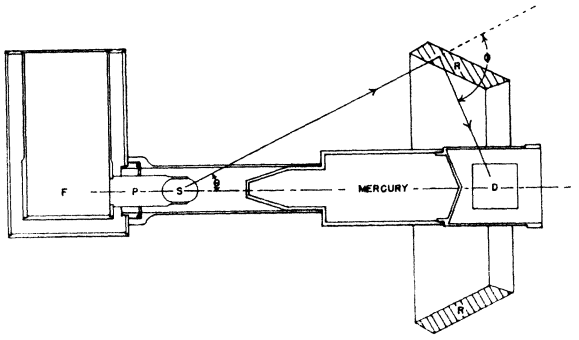

FIG. 1. Experimental apparatus.

the expression[7]

$$\bar{\sigma}=\frac{(2j_1+1)}{(2j_0+1)}\frac{h^2c^2\Gamma}{4E_0^2}\frac{N(E_{\rm res})}{N}A_d, \tag{1}$$

where j_1 is the spin of the excited nuclear state through which scattering is taking place, j_0 is the spin of the nuclear ground state, E_0 is the excitation energy of the excited state, and Γ is the level width of the excited state. Since the lifetime τ of the excited state is $\tau=\hbar/\Gamma$, it is given by the expression

$$\tau=\frac{gh^3c^2}{8\pi E_0^2\bar{\sigma}}\frac{N(E_{\rm res})}{N}A_d, \tag{2}$$

where $g=(2j_1+1)/(2j_0+1)$.

The procedure used in determining $\bar{\sigma}$ will be discussed in Sec. IV and the calculation of $N(E_{\rm res})/N$ is treated in Sec. VI.

II. EXPERIMENTAL APPARATUS

The scattering apparatus is shown in Fig. 1. The gamma detector D is a 1.5 in. high by 1.5 in. in diam NaI(Tl) crystal coupled to an RCA 6655 photomultiplier tube. This scintillation counter feeds its output pulses into a single-channel differential pulse-height selector. The source S is contained in a Pyrex glass ampule which is held in place, under spring pressure, in a copper prod P making good thermal contact with the inner container of a metal Dewar flask F. In this way it is possible to control the temperature of the arsine source gas and, when desired, freeze it by placing liquid nitrogen in the Dewar. The thin aluminum tube connecting the outer Dewar container with the mercury-filled shield makes it possible to evacuate the region around the source and helps maintain a rigid source geometry. The mercury shield, which is 19 cm in length, attentuates direct gamma radiation from the source to the counter by a factor of greater than 10^6. The scintillation counter is rigidly attached to the thin steel shell surrounding its crystal D. Three aluminum support arms extending from the base of the scintillation counter hold the scattering ring R in place. The entire apparatus has cylindrical symmetry and was supported in the middle of a room by eight 80-mil stainless steel wires to minimize background scattering.

The two scattering rings were both cast metallic rings split along a diameter so that they could be removed and interchanged. It was necessary to anneal the selenium ring for approximately 5 h at 218°C to get a complete transition from the glassy to the metallic state. In the glassy state, the selenium ring would not hold its shape over long periods. Both the selenium ring and the comparison zinc ring were x rayed with a CO^{60} source in order to be sure that the castings were uniform. They were also checked for density. Both appeared to be free of voids.

III. THE SOURCE AND ITS PREPARATION

As mentioned earlier, the source used in this work was in the form of arsine gas (AsH_3) at a pressure of less than 2 mm of Hg. The establishment of the resonance condition dictated the use of a gaseous source at low pressure so that the nucleus which emits a gamma ray will be reasonably free to recoil from the previous beta decay until the gamma is emitted. If $N(E_{\rm res})/N$ is to be calculated with any reliability, the recoil between beta decay and gamma emission must involve negligible exchange of momentum of the recoiling nucleus with other systems. A source in the form of arsine gas at a few millimeters of pressure comes very close to satisfying this last requirement. Since most of the molecular mass resides in the arsenic atom, only a small fraction of the recoil energy will be available in the center-of-mass system for molecular decomposition. Due to this fact, there is good reason to believe that in greater than 90% of the cases the beta decay does not lead to molecular breakup until after the gamma ray has been emitted. In those cases where breakup occurs, only a small fraction of the nuclear recoil momentum will be lost to the hydrogen atoms. By keeping the gas pressure below a few millimeters of mercury, molecular collisions between beta decay and gamma emission will be negligible.

Two precautions were found to be very important in the source preparation. It was necessary that great care be taken in cleaning the source ampule if decomposition of the arsine was not to be a factor in the experiment. It was also important that the arsine be of very high purity with all traces of moisture removed. This was accomplished by passing the arsine through a dry ice-acetone trap. The gas was then triple distilled through a series of liquid nitrogen traps, all of which were preflushed with hydrogen. After the distillation, the arsine was frozen in the source ampule and all residual gasses pumped off. Then, with the arsine still at liquid nitrogen temperatures, the ampule was sealed off with a torch. With this procedure, inactive arsine ampules have been kept, away from light, for over six months without showing any sign of decomposition.

The sources used in this work were prepared from 20 mC shipments of $AsCl_3$ in acid solution, obtained from Oak Ridge. The chemical yield was approximately 93%.

IV. DETERMINATION OF THE AVERAGE CROSS SECTION

The basic method used in determining the average scattering cross section for resonant scattering through the 0.559-MeV first excited level of Se^{76} depended upon the use of zinc and selenium scattering rings which gave essentially the same scattering for all events, exclusive of resonant scattering, which could contribute to the detector counting rate. The difference in the counting rates of the detector when the two scattering rings are interchanged should be due to resonant scattering.

Consideration of the scattering geometry shows that the counting rate N due to resonant scattering is given by the expression

$$N=\frac{n\bar{\sigma}}{4\pi}\int\left\{W(\phi)(I\eta d\Omega_c/4\pi)\int_{r_0(\theta)}^{r_m(\theta)}e^{-(\lambda\Delta r+\lambda'\Delta r')}dr\right\}d\Omega_s, \quad (3)$$

where n is the number of scattering atoms per unit volume, I is the total number of 0.559-MeV photons emitted per second by the source, η is the average counter detection efficiency, $d\Omega_c$ is the solid angle subtended by the counter at a point in the scatterer, Ω_s is the solid angle subtended at the source by the scattering ring, θ is the angle between the system axis and a line running from the source to a scattering point in the scattering ring, Δr is the distance which a photon must penetrate into the scattering ring to reach the scattering point and is a function of r and θ, $\Delta r'$ is the distance which a scattered photon must travel through the scattering ring in going from the scattering point to the counter and is also a function of r and θ, λ is the good geometry linear absorption coefficient of the scattering material for an entering photon and was taken as $(1.045\times10^{-23}$ cm^2/atom$)\times n$ for selenium, λ' is the poor geometry linear absorption coefficient for photons leaving the scattering ring, $W(\phi)$ is the angular dependence of the resonant scattering process and is normalized so that $\int W(\phi)d\Omega=4\pi$, ϕ is the angle of scattering and is a function of θ, r_0 is the distance from the source to the point of entrance of a photon into the scattering ring and is a function of θ, and r_m is the distance from the source to the point where a photon would leave the scattering ring if not scattered. The function $C(\theta)=I\eta d\Omega_s/4\pi$ and λ' are determined experimentally, and the integrals are performed numerically. The measured value of λ' was $(0.768\pm0.005)\lambda$. With this expression, $\bar{\sigma}$ may be computed from N.

The experimental determination of $C(\theta)$ is made by noting that it is just the counter counting rate which would be observed if the source were located at the scattering point. By placing a point source at various points in the region to be occupied by the scattering ring and observing the counting rate, $C(\theta)$ may be determined within a constant factor. This factor is the ratio of the strength of the scattering source to the survey source.

In practice, it is very difficult to balance the zinc and selenium scattering rings exactly, therefore, the ratio of the scattering from the selenium and zinc rings in the absence of resonant scattering must be known. This was determined both experimentally and theoretically. In the theoretical calculation, it was assumed that three processes other than background and resonance fluorescence contributed to the counting rate. They were Thompson scattering, Rayleigh scattering, and unresolved annihilation radiation from positrons produced in the scattering ring by pair production due to higher energy gamma rays. The experimental ratio was determined using three different sources for which the resonance condition should have been destroyed by collision of the recoiling nucleus with its neighbors. One source was a solid As_2O_3 source. Another source was an HCl solution of $AsCl_3$. The final source was one of the arsine sources which was frozen with liquid nitrogen in the Dewar supporting the ampule. The three experimental ratios and the theoretical ratio all agreed within experimental error.

V. EXPERIMENTAL RESULTS

As indicated earlier, the experimental information needed for a calculation of the average resonance scattering cross section is a determination of the function $C(\theta)$, and the ratio of the counting rate with the selenium scattering ring in place to that with the zinc ring in place under both resonant and nonresonant conditions. In the actual experiment it was not $C(\theta)$ that was determined but the function $N(\theta)=C(\theta)/C(23.5°)$. After $N(\theta)$ was once determined, the $C(\theta)$ could be obtained for a given scattering source by measuring the ratio R of its strength to that of a smaller point comparison source whose counting rate C_S at the 23.5° position is also determined. Then $C(\theta)=C_SRN(\theta)$. Figure 2 is a plot of the measurements of $N(\theta)$ for the one geometry which was kept fixed for all of the measurements being reported here. In the case of the As^{76} to Se^{76} decay, special consideration must be given to the experimentally determined value of C. As a study of the currently accepted decay scheme (Fig. 3) for As^{76} shows, there is in addition to the 0.559-MeV gamma ray from the first excited state to the ground state of Se^{76}, another gamma ray of energy 0.56 MeV. These two gamma rays will be unresolved by the scintillation counter, but the second gamma cannot contribute to the resonant scattering. For this reason the experimental value of C_S must be corrected to bring it down to the value which is due to the ground state gamma alone.

For the determination of the difference in scattering

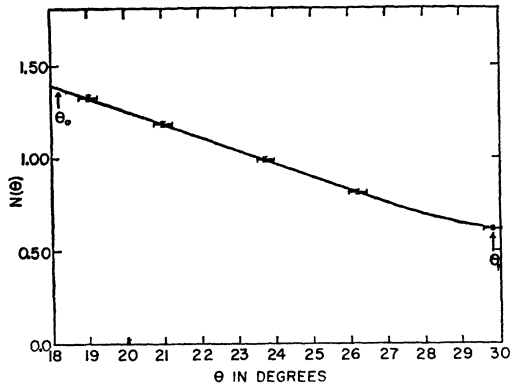

FIG. 2. The function $N(\theta)$ representing the detection efficiency of the scintillation counter for various source positions located on a surface at constant depth within the scattering ring. These efficiencies are measured relative to the detection efficiency for the source in the 23.5° position. Within the experimental uncertainty, $N(\theta)$ is not changed as the depth of the surface within the scattering ring is varied.

between the selenium and zinc scattering rings, the same procedure was used under both resonant and non-resonant conditions. The window width of the scintillation spectrometer was first set approximately equal to the full width at half maximum of the photo peak of the 0.559-MeV Se^{76} gamma ray, and the center of the window was positioned at the peak. This adjustment was checked at hourly intervals throughout a run. Each time this check was made, background counts were taken, and the survey source mentioned above was counted as a check on detector efficiency. Between

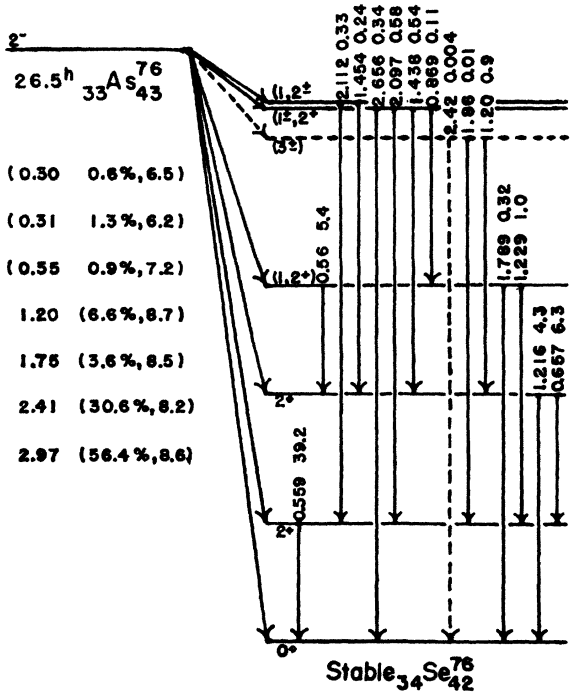

FIG. 3. The $As^{76} \rightarrow Se^{76}$ decay scheme as taken from the National Research Council Nuclear Data project sheet NRC 59-5-36.

these readings, the zinc and selenium scattering rings were alternated four or five times with eight to ten minute counts being taken on each. Table I shows a representative sample of the data (corrected for background and decay) taken on one of the runs with a gaseous arsine source. For this run $C_S = (5.70 \pm 0.01) \times 10^5$ min^{-1} and $R = 8.75 \pm 0.02$. Each individual count involved a total of 2 560 counts. Similar data were taken using a solid As_2O_3 source and also one containing As_2Cl_3 in HCl solution. During part of another run with a gaseous arsine source, the source was frozen with liquid nitrogen. With each of the two solid sources and the liquid one the possibility of resonance should have been destroyed by collisions within the source. In each of the three cases, the ratio of the selenium to zinc counting rate was the same, and therefore the results from the three cases were combined to obtain the selenium to zinc ratio in the absence of resonance. This value is

$$(\mathrm{Se/Zn})_{\mathrm{nonres}} = 0.927 \pm 0.004. \quad (4)$$

In each case with the arsine source, the counting rate due to resonance scattering was taken as

$$N = \mathrm{Se}_{\mathrm{res}} - (\mathrm{Se/Zn})_{\mathrm{nonres}} \mathrm{Zn}_{\mathrm{res}}. \quad (5)$$

In the resonant and nonresonant cases, the fluctuations in both the individual selenium and the individual zinc counting rates for a given run extending over several days were just that which should have been expected due to counting statistics. For the run shown in Table I, $N = 57.3 \pm 3.5$ counts/min.

The average value of $\bar{\sigma}$ obtained from the data is

$$\bar{\sigma} = (6.33 \pm 0.45) \times 10^{-27} \text{ cm}^2. \quad (6)$$

In obtaining this value for $\bar{\sigma}$, the results of two of the early arsine runs were not included because of the large variation within the data in these runs. This variation was due to two causes. First, if great care is not taken in cleaning the source ampule and purifying the arsine, the arsine source will decompose with the deposit of metallic arsenic on the walls. This requires corrections to the data which are difficult to make. The second problem is drift in calibration of the apparatus. If a lead screen around the counter crystal is not used for preferential screening, the very large counting rate due to low-energy Compton scattered radiation can cause serious drifting in the photomultiplier gain as a result of fatigue in the last few dynodes. A half centimeter lead shield around the counter, along with reduced photomultiplier voltage and greatly improved calibration techniques, completely eliminated this problem in the later runs. Figure 4, showing a limited region of the pulse-height spectrum of the scattered radiation from both a selenium and zinc scattering ring using an arsine source, shows why small drifts in gain in the apparatus can be more serious than would be the case if the photopeak of the scattered radiation were pronounced

TABLE I. Sample of data taken under resonant scattering conditions: The tabulated counting rates have been corrected for both decay and background. They were collected over an interval of about three days. The time sequence of taking these data was such that a given counting rate from the selenium scatterer was taken prior to that recorded on the same line for the comparison zinc scatterer and following that recorded on the previous line for zinc.

Counts/min		Counts/min	
Se	Zn	Se	Zn
204.3	166.8	215.9	155.2
221.7	168.1	209.0	169.2
209.0	167.1	205.2	171.0
214.2	191.9	206.8	176.9
216.3	172.0	226.6	166.6
221.2	172.8	222.0	167.8
204.4	150.4	202.3	137.7
235.6	172.0	211.3	156.5
189.9	168.8	219.9	159.4
215.0	173.3	219.1	176.3
215.4	172.9	234.2	172.4
213.5	146.1	189.7	203.3
		Se av = 213.5	Zn av = 168.4

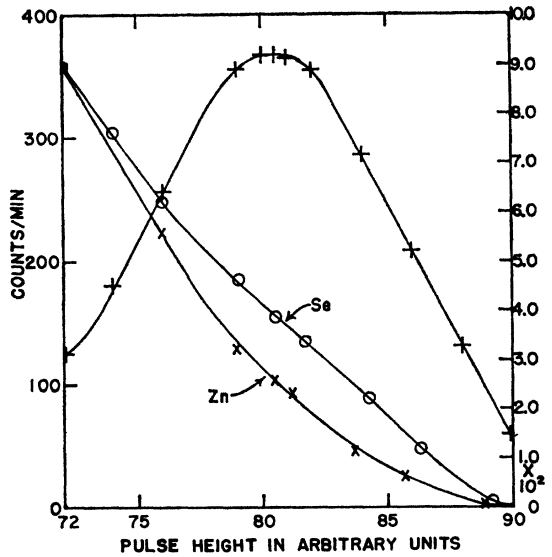

FIG. 4. Counting rate as a function of pulse height for a limited region of the pulse-height spectrum of the scattered radiation. The region shown is around the position of the photopeak of the 0.559-MeV gamma-ray line from Se^{76}. The open circles represent the counting rate from a gaseous arsine source with the selenium scatterer in place. The ×'s represent the counting rate with a zinc scatterer substituted for the selenium one. The left-hand counting rate scale applies to these curves. The +'s represent the counting rate for direct radiation from the small As^{76} survey source and show the 0.559-MeV photopeak for comparison. The right-hand counting rate scale applies to this curve.

enough to produce a local maximum. The data for Fig. 4 were taken in one of the two early runs mentioned above and show slight signs of apparatus drift.

VI. THE CALCULATION OF $N(E_{res})/N$

The calculation of $N(E_{res})/N$, in essence, involves a consideration of two effects. First, it is necessary to know the fraction of nuclei recoiling from the beta-decay process which will have the necessary component of recoil momentum along a direction from the source to the scatterer. Then it is necessary to know what fraction of these nuclei will emit a gamma ray in the desired direction. The fraction of the recoiling nuclei which will have the desired recoil momentum component will depend upon the nature of the beta-neutrino angular correlation. The probability of gamma emission in the desired direction by this selected group of recoiling nuclei will depend upon the degree of spin polarization of this group and the multipolarity of the gamma ray. This group of nuclei may have a net spin polarization even for an allowed beta-decay process. The possibility of such a polarization may be understood by considering the beta-neutrino emission process. The probability of production of a beta and neutrino giving the desired recoil momentum is proportional to nuclear matrix elements which are not averaged equally over all neutrino directions associated with any given energy and direction for the beta particle. Therefore, the contributions from the various changes, Δm, in nuclear magnetic quantum number are, in general, not equal.

In the case of resonant scattering through the first excited state of Se^{76}, contributions to $N(E_{res})/N$ come not only from the simple process of a beta-decay followed by the resonant gamma ray, but some significant contribution is also made by processes in which the beta decay is followed by one or two intermediate gamma rays before the resonant gamma ray is emitted. Morita, Morita, and Yamada[8] derived expressions for the analysis of such cases when the beta-decay process is allowed; however, no such analysis has been reported in the literature for cases in which the beta-decay process is forbidden. The author has carried out a theoretical investigation of the various resonance fluorescence experiments which might be used to aid in obtaining information about the nuclear matrix elements in some interesting forbidden beta-decay processes. In this work, which will be published later, it is shown that for any beta-decay process followed by n gamma rays the expression for the probability of emission of the beta particle, neutrino, and following gammas in the directions of the unit vectors ω_β, ω_ν, $\omega_1, \omega_2, \cdots, \omega_n$ which are contained within the respective solid angles $d\Omega_\beta, d\Omega_\nu, d\Omega_1, d\Omega_2, \cdots, d\Omega_n$ is proportional to

$$4\sum F(Z,E)\,[(2k_a+1)(2j_a+1)]^{1/2}(2j_f+1)^{-1/2}[(\alpha_a j_a|\rho_a|\alpha_a j_a)]_{q_a}{}^{k_a}[(\alpha_f j_f|\rho_f|\alpha_f j_f)]_{q_f}{}^{k_f}\mathfrak{D}_{\mu_a q_a}{}^{(k_a)-1}(\mathbf{a})\mathfrak{D}_{\mu_f q_f}{}^{(k_f)}(\mathbf{f})$$
$$\times\{\sum(-1)^{k_\beta-q_\beta}(2k_\beta+1)^{-1/2}b_{k_\beta}(L_\beta L_\beta';\, k_e k_\nu, q_e q_\nu)\bar{F}_{k_\beta}(L_\beta L_\beta';\, j_a j_b;\, k_a k_b)Q_{k_\beta q_\beta}(k_e k_\nu;\, q_e q_\nu)H_{k_a\mu_a}(k_\beta q_\beta;\, k_b q_b)\}$$
$$\times\{\sum(\tau_1)^{k_1}G(L_1 L_1' j_b)H_{k_b q_b}(k_1\mu_1;\, k_c q_c)\bar{F}_{k_1 q_1}(L_1 L_1';\, j_c j_b;\, k_c k_b)\mathfrak{D}_{\mu_1 q_1}{}^{(k_1)-1}(\omega_1)\}\cdots\{\sum(\tau_n)^{k_n}G(L_n L_n' j_{n-1})$$
$$\times H_{k_{n-1}q_{n-1}}(k_n\mu_n;\, k_f\mu_f)\bar{F}_{k_n q_n}(L_n L_n';\, j_f j_{n-1};\, k_f k_{n-1})\mathfrak{D}_{\mu_n q_n}{}^{(k_n)-1}(\omega_n)\}d\Omega_\beta d\Omega_\nu d\Omega_1\cdots d\Omega_n, \quad (7)$$

[8] M. Morita, R. S. Morita, and M. Yamada, Phys. Rev. 111, 237 (1958).

where the first summation is over q_a, k_a, k_b, k_c, $\cdots$, k_{f-1}, k_f, k_e, k_ν, μ_a, q_b, q_c, $\cdots$, q_{f-1}, q_f, and μ_f; the second summation is over k_β, q_β, q_e, q_ν, L_β, and L_β'; and the third and succeeding summations are over k_1, μ_1, L_1, L_1', q_1; k_2, μ_2, L_2, L_2', q_2; $\cdots$; and k_n, μ_n, L_n, L_n', q_n. Also

$$G(L_iL_ij_s)=\delta_{L_i}\delta_{L_i}(2j_s+1)^{-1},$$

$$H_{k_1q_1}(k_\gamma\mu_\gamma;\,k_2q_2)=(-1)^{k_1-q_1}[(2k_\gamma+1)(2k_2+1)]^{1/2}\bar{V}(k_\gamma k_2k_1;\,\mu_\gamma q_2-q_1),$$

$$Q_{k_\beta q_\beta}(k_ek_\nu;\,q_eq_\nu)=[(2k_e+1)(2k_\nu+1)]^{1/2}\sum(-1)^{k_e+k_\nu}\bar{V}(k_ek_\nu k_\beta;\,\mu_e\mu_\nu-q_\beta)\mathfrak{D}_{\mu_eq_e}^{(k_e)-1}(\omega_\beta)\mathfrak{D}_{\mu_\nu q_\nu}^{(k_\nu)-1}(\omega_\nu)$$

with the summation over μ_e and μ_ν,

$$\bar{F}_{k_\gamma q_\gamma}(L_\gamma L_\gamma';\,j_cj_b;\,k_ck_b)=(\tau)^{k_\gamma}(-1)^{L_\gamma+L_\gamma'}[(2j_b+1)(2j_c+1)(2k_\gamma+1)]^{1/2}\times X(L_\gamma j_cj_b/L_\gamma' j_cj_b/k_\gamma k_ck_b)[(\alpha_\gamma L_\gamma|\rho_\gamma|\alpha_\gamma' L_\gamma')]_{q_\gamma}^{(k_\gamma)},$$

$$\bar{F}_{k_\beta}(L_\beta L_\beta';\,j_bj_a;\,k_bk_a)=(-1)^{L_\beta-1}(2k_\beta+1)[(2j_a+1)(2j_b+1)(2L_\beta+1)(2L_\beta'+1)]^{1/2}\times\bar{V}(L_\beta L_\beta' k_\beta;\,1\;-1\;0)X(L_\beta j_bj_a/L_\beta' j_bj_a/k_\beta k_bk_a),$$

and

$$b_{k_\beta}(L_\beta L_\beta';\,k_ek_\nu,q_eq_\nu)=\tfrac{1}{4}\sum[F(Z,E)(2j_a+1)(2L_\beta+1)^{1/2}(2L_\beta'+1)^{1/2}\bar{V}(L_\beta L_\beta' k_\beta;\,1\;-1\;0)]^{-1}(-1)^{L_\beta'+1-k_e-k_\nu}\times X(j_ej_\nu L_\beta/j_e'j_\nu' L_\beta'/k_ek_\nu k_\beta)B_\epsilon^{L_\beta*}(\kappa_e,\kappa_\nu)B_{\epsilon'}^{L_\beta*}(\kappa_e',\kappa_\nu')[(\alpha_ej_e|\rho_e|\alpha_e'j_e')]_{q_e}^{(k_e)}[(\alpha_\nu j_\nu|\rho_\nu|\alpha_\nu'j_\nu')]_{q_\nu}^{(k_\nu)}$$

with the summation over ϵ, ϵ', κ_e, κ_e', κ_ν, and κ_ν'. In this expression the $[(\alpha_aj_a|\rho_a|\alpha_aj_a)]_{q_a}^{(k_a)}$ and $[(\alpha_fj_f|\rho_f|\alpha_fj_f)]_{q_f}^{(k_f)}$ represent elements of the irreducible matrix representations of the density matrices of the observed initial and final nuclear states according to the notation of Fano and Racah.[9] Likewise the $[(\alpha_ej_e|\rho_e|\alpha_e'j_e')]_{q_e}^{(k_e)}$, $[(\alpha_\nu j_\nu|\rho_\nu|\alpha_\nu'j_\nu')]_{q_\nu}^{(k_\nu)}$, and $[(\alpha_\gamma L_\gamma|\rho_\gamma|\alpha_\gamma' L_\gamma')]_{q_\gamma}^{(k_\gamma)}$ represent elements of the density matrices for the observed electron, neutrino, and gamma ray states. The δ_L are the reduced nuclear matrix elements for the gamma-ray transitions of multipole order L. The $F(Z,W)$ is the usual Fermi function containing the energy dependence and Coulomb correction factor for allowed beta spectra. The $\mathfrak{D}_{\mu_jq_j}^{(k_j)}(\omega_j)$ are elements of the irreducible rotation matrix which rotates from the laboratory coordinate system to the one in which the density matrix for the particle, gamma ray, or nuclear state j is constructed. In the case of radiation, this latter coordinate system is chosen with its Z axis along the direction of the radiation. The remaining notation is the same as that used by Fano and Racah.[9] It should be noted that if no transverse polarization measurements are made on the beta or neutrino the only elements in their density matrices that are different from zero are those for which $q_e=q_\nu=0$. In this case the particle parameters $b_{k_\beta}(L_\beta L_\beta;\,k_ek_\nu;\,00)$ and the geometrical factors $Q_{k0}(k_ek_\nu;\,00)$ reduce to those which are tabulated by Bincer.[10] In using these tables care must be taken since there are a few sign errors.

With the use of Eq. (7), it is possible to compute the probability that the final or nth gamma ray will be emitted within a given solid angle with its energy falling within unit energy range around any given energy. This is done by noting that to the necessary accuracy, the energy E of this gamma ray will be given by

$$E=E_0(1-E_0/2Mc^2+P/Mc),\tag{14}$$

where E_0 is the excitation energy of the level emitting this gamma ray, M is the mass of the emitting nucleus, and P is the momentum component of the emitting nucleus along the nth gamma ray just prior to emission of this gamma ray. The momentum of the emitting nucleus is due to the resultant recoil from the beta, neutrino, and all intermediate gamma rays. It is assumed, as is also the case in Eq. (1), that the maximum recoil energy due to these causes is roughly an order of magnitude larger than the thermal energy of the source and scattering atoms. Since this gives

$$dE=(E_0/1822A)dP,\tag{15}$$

with P now being expressed in units of m_0c and A being the mass number of the nucleus, it is clear that the probability that the nth gamma ray will have an energy falling within the interval between E and $E+dE$ and be directed within the solid angle $d\Omega_n$ is just $E_0/1822A$ times the probability that the resultant nuclear recoil momentum component P will fall between P and $P+dP$ while gamma n is contained within $d\Omega_n$. This latter probability is obtained from Eq. (7) by integrating this expression over all combinations of angles (except those involved in $d\Omega_n$) and beta energies that are consistent with the condition

$$-P\gtrless[(W^2-1)^{1/2}\omega_\beta+(W_0-W)\omega_\nu+\sum_{i=1}^{n-1}E_i\omega_i]\cdot\omega_n\gtrless-(P+dP),\tag{16}$$

where W is the total energy (kinetic+rest mass) of the beta particle, W_0 the maximum beta energy, and E_i the energy of the ith gamma ray, all in units of m_0c^2. The resulting multiple integral will contain a series of terms, from each of which may be factored a reduced

[9] U. Fano and G. Racah, *Irreducible Tensorial Sets* (Academic Press Inc., New York, 1959).

[10] Adam M. Bincer, Phys. Rev. **112**, 244 (1958).

nuclear matrix element. Once this has been done, each of the individual terms may be integrated numerically and the overall expression evaluated after some assumption is made concerning values for the ratios of the various nuclear matrix elements. The numerical integrations with the Coulomb factor included were done using an IBM 1620 computer.

The value of $N(E_{res})/N$ for the 0.559-MeV gamma-ray line of Se^{76} resulting from a free recoil decay of As^{76} was obtained by carrying out the type of calculation outlined above on each decay path contributing significantly to this line. The choice of the nuclear matrix element ratios for the beta groups leading to each decay path is simplified by the fact that for each of these paths there is one value of $|P|$ for which the desired integral becomes independent of the ratios of the nuclear matrix elements. This means that for any decay path for which this special value of $|P|$ is close to the P which produces resonance, no knowledge of the nuclear matrix elements is needed. In our case, this condition holds well enough to make the uncertainty due to lack of knowledge of nuclear matrix elements small for all but the most important group contributing to the 0.559-MeV line. This is the 2.41-MeV first forbidden beta group leading directly to the 0.559-MeV level.

In the indicated calculation of $N(E_{res})/N$, it has been assumed that the beta interaction is of the vector-axial vector type. The simplification of neglecting all but the $\xi^2=(\alpha Z/2R)^2$ terms has been made when dealing with the forbidden beta contributions. This approximation has no significant effect upon the final answer except for its possible effect upon the contribution from the 2.41-MeV beta group. For this group, the ξ^2 approximation might be justified on the basis of the lack of any noticeable departure of the energy spectrum shape from the statistical one.[11] On the other hand, the presence of a beta-gamma angular correlation[12] and its large $f\tau$ value suggest that this approximation may not be too good. In any case the effect is not expected to be large enough to justify the extra labor involved in including higher order terms for this experiment.

The beta and gamma-ray branching ratios used in these calculations are those reported in the National Research Council Nuclear Data Sheets NRC 59-5-39 and NRC 59-5-40 on As^{76}. The value of $N(E_{res})/N$ calculated in this way may be expressed as

$$N(E_{res})/N=(0.0269\pm0.0008)-0.0033A, \quad (17)$$

where A is a parameter depending upon the matrix elements for the 2.41-MeV beta process. The uncertainty indicated in the first term on the right of (17) represents that due to a complete uncertainty in the nuclear matrix elements associated with all beta groups except the 2.41-MeV one. The parameter A is given by the expression

$$A=(1-\tfrac{2}{3}|X|^2)/(1+|X|^2), \quad (18)$$

with

$$X=\frac{C_A|\int\boldsymbol{\sigma}\times\mathbf{r}/r|+C_V|\int\mathbf{r}/r|}{C_A|\int\boldsymbol{\sigma}\cdot\mathbf{r}/r|}. \quad (19)$$

It has been assumed that $C_A=C_A'$ and $C_V=C_V'$ are real, as seems to be justified by experimental results. The fact that the factor multiplying $|X|^2$ in the numerator of (18) is 2/3 rather than 1/3 represents the effect on the direction of emission of the gamma rays of the spin polarization of the nuclei recoiling from the beta decay.

VII. DISCUSSION

An expression for the lifetime $\tau_{0.559}$ of the 0.559-MeV level of Se^{76} is obtained by inserting the experimental value of $\bar{\sigma}$ from (6) and the calculated value of $N(E_{res})/N$ from (17) into (2). This gives

$$\tau_{0.559}=(1.55\pm0.12)-(0.19\pm0.01)\frac{1-\frac{2}{3}|X|^2}{1+|X|^2}\times10^{-11}\text{ sec}. \quad (20)$$

The indicated uncertainties now include the experimental ones. From (20) it is seen that $\tau_{0.559}$ can be as small as 1.36×10^{-11} sec or as large as 1.68×10^{-11} sec depending upon the value of $|X|^2$. The circular polarization correlation measurement of Boehm[12] and the nuclear orientation work of Pipkin and Culvahouse[13] give some indication of the value of $|X|^2$. The X used here corresponds (to the approximation of keeping only the highest power of ξ) to that used by Boehm. In the notation of Pipkin and Culvahouse, their $\alpha_1/\alpha_2=|X|^2$. Boehm's work indicates that $|X|^2$ should be close to 1.6×10^{-3} if X is positive, whereas $|X|^2$ should be close to 36 if X is negative. The results of Pipkin and Culvahouse would tend to favor the smaller of Boehm's two values, as would the theoretical arguments of King and Peaslee.[14] If this choice is approximately correct, $\tau_{0.559}=(1.36\pm0.13)\times10^{-11}$ sec. On the other hand, there are two values of τ available from Coulomb excitation experiments.[15,16] The work of Temmer and Heydenburg gives $(1.7\pm0.2)\times10^{-11}$ sec, whereas that of McGowan and Stelson gives $(1.23\pm0.11)\times10^{-11}$ sec. The smaller value of $|X|^2$ gives reasonable agreement with McGowan and Stelson, but is not quite consistent with the result of Temmer and Heydenburg. If one were to accept their result in preference to that of McGowan and Stelson, then Boehm's larger choice for $|X|^2$ would be indicated. Choosing $|X|^2=33$ gives $\tau_{0.559}=(1.67\pm0.13)\times10^{-11}$ sec. This value is in excellent agreement with Temmer and Heydenburg.

[11] A. V. Pohm, R. C. Waddell, and E. N. Jensen, Phys. Rev. **101**, 1315 (1958).
[12] F. Boehm, Z. Physik **152**, 384 (1958).
[13] F. M. Pipkin and J. W. Culvahouse, Phys. Rev. **109**, 1423 (1958).
[14] R. W. King and D. C. Peaslee, Phys. Rev. **94**, 1284 (1954).
[15] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **104**, 967 (1956).
[16] F. K. McGowan and P. H. Stelson, Oak Ridge National Laboratory Report ORNL-2610, 1959 (unpublished), p. 11.

The above comparisons might be changed, however, if an appreciable contribution is made by the beta-decay mode in which the beta particle and neutrino carry off two units of total angular momentum (the B_{ij} matrix element). The dependence indicated by (17) of $N(E_{\text{res}})/N$ on the nuclear matrix elements would, in this case, also include a dependence upon $|B_{ij}|^2$ and interference terms with it. The arguments of King and Peaslee[14] would suggest that such a contribution by the B_{ij} matrix element might not be unexpected in the 2.41-MeV beta decay of As^{76}. The form of the contributions from the B_{ij} matrix element to $N(E_{\text{res}})/N$ make it seem unlikely, however, that the extreme range of the matrix element dependent part of $N(E_{\text{res}})/N$ would be much different than it is now, and the first term in (17) would be unchanged.

When one considers factors which could change the value of $\tau_{0.559}$ obtained in this work, it should be noted that any inaccuracy in the As^{76} to Se^{76} decay scheme or branching ratios could have a serious effect. Any increase in the relative intensity of gamma rays whose photopeaks can not be resolved from that of the 0.559-MeV gamma ray would lead to a reduction in the quoted value of $\tau_{0.559}$ since, in effect, it would mean that too high a value of $N(E_{\text{res}})/N$ was used. A large change in the intensities of the higher energy gamma rays would change the correction which is made to $C(\theta)$ for the effect on the counting rate, in the region of the 0.559-MeV photopeak, of the Compton distributions from these higher energy gamma rays. In practice this correction, along with that required by the presence of the 0.56-MeV gamma ray, was made on the calculated value of $N(E_{\text{res}})/N$ rather than on the experimental value of $C(\theta)$ since both corrections may be made most easily in this way.

In addition to the measurements made with low-pressure arsine sources, one run was made with an arsine source at approximately four atmospheres of pressure to determine what effect the increased density would have upon $N(E_{\text{res}})/N$. At this pressure, $N(E_{\text{res}})/N$ was reduced to about 81% of its value at a source pressure of less than 2 mm of mercury. An attempt was also made with a low-pressure arsine source to look for scattering through the 1.216-MeV level of Se^{76}. No evidence was seen of any resonant scattered counts at this energy, but the counting statistics were such that the only thing which can be said with any reliability is that the number of resonent scattered counts was less than 3 per min. If the level width of this state were the same as that for the 0.559-MeV level, the resonant scattered counting rate should be expected to be about 1% of that from 0.559-MeV level. This is due to a factor of about 0.10 in the ratio of gamma-ray intensities, one of about 0.5 due to the difference in apparatus detection efficiencies at the two energies, and one of about 0.2 due to the energy dependence of $\bar{\sigma}$. Therefore, a resonant scattering counting rate of 0.57 count per min for the run listed in Table I would have implied a lifetime of about 1.6×10^{-11} sec for the 1.216-MeV level. Since the counting rate was less than 5 (i.e., 3/0.57) times that to be expected for such a lifetime, the value of $\tau_{1.216}$ must be greater than 1/5 that of the 0.559-MeV level. If one corrects further for the fact that $N(E_{\text{res}})/N$ for the 1.216-MeV level is about 7/8 that for the 0.559-MeV level, then $\tau_{1.216}>4\times10^{-12}$ sec.

This work shows the extent to which a lifetime measured by a resonant scattering experiment of the type discussed in this paper is dependent upon a knowledge of the nuclear matrix elements for the beta-decay process. In fact, the results reported here suggest that it would be valuable to carry out further measurements on Se^{76}. In these measurements, several things should be done. First, arrangements should be made for automating the exchange of the selenium and comparison zinc scatterers. This would allow the use of sources of greater intensity and provide an even more precise scattering geometry. Second, the use of a multichannel pulse-height analyzer would improve the accuracy with which the value of $C(\theta)$ can be corrected for the effects of higher energy gamma rays. It would also make it possible to investigate both the 0.559- and the 1.216-MeV level at the same time. In addition, the great care required in continuous energy calibration of the equipment could be met more easily with resultant increase in the data taking duty cycle. Third, not only a resonance scattering experiment but also a resonance absorption experiment should be done with the same geometry. This would allow a determination of τ from the absorption experiment and of $\bar{\sigma}$ from the scattering experiment. A combination of these results would give a determination of $N(E_{\text{res}})/N$ which could be used for an evaluation of the beta-decay matrix element ratio $|X|^2$. If such data were available, then a more careful calculation of $N(E_{\text{res}})/N$ including the contribution of the B_{ij} matrix element and, perhaps, terms of the order of ξ in the other matrix elements would be justified. A combination of results from these experiments with a beta resonant gamma coincidence experiment could lead to a reasonably accurate evaluation of some of the matrix element ratios for the 2.41-MeV beta decay of As^{76}. The first part of the suggested work along with similar measurements on the first excited state of tellurium-124 are currently under way as a Ph.D. dissertation at Bryn Mawr, and preparations are also being made for the beta resonant gamma coincidence experiment.

ACKNOWLEDGMENTS

The author would like to express his thanks to Dr. Franz Metzger of the Bartol Research Foundation for several helpful discussions. A particular debt of gratitude is owed to Dr. A. L. Patterson of The Institute for Cancer Research for his generosity in allowing extensive use of his IBM 1620 computer prior to the acquisition of our own. Thanks are due to Professor Walter C. Michels for some helpful comments on the manuscript.