Nuclear Resonance Fluorescence in Ge^{72} and Ge^{74}†

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With gaseous sources of As^{72} and As^{74}, large nuclear resonance fluorescence effects have been observed for the 835-kev excited state of Ge^{72} and for the 596-kev excited state of Ge^{74}. The angular distribution of the resonance radiation characterizes the spins of both these excited states as 2 and the spins of the ground states as 0.

From the magnitude of the resonance fluorescence effects, the mean lives of the 835-kev and 596-kev gamma-ray transitions were determined as $(4.6\pm1.2)\times10^{-12}$ second and $(1.9\pm0.3)\times10^{-11}$ second, respectively. These lifetimes are approximately one order of magnitude shorter than those expected for electric quadrupole transitions on the basis of the single particle model.

The influence of collisions in the gas on the observed resonance fluorescence effect was studied using the 596-kev level in Ge^{74}.

INTRODUCTION

THE study of nuclear resonance fluorescence provides us with very useful information concerning the transition probabilities of fast gamma-ray transitions whose lifetimes are beyond the range of the delayed coincidence method, i.e., shorter than 10^{-10} to 10^{-11} sec. The sensitivity of the resonance fluorescence method increases with decreasing lifetime, i.e., with increasing width. Although only transitions leading to the ground states of stable nuclei can be studied, the knowledge of the branching between competing gamma-ray transitions will allow one in some cases to draw conclusions concerning transitions between excited states.

In addition to the information concerning transition probabilities obtained from the magnitude of the resonance fluorescence effect, the study of the angular distribution of the resonance radiation represents a very direct way of determining spins of excited states, multipole orders of transitions, and mixing ratios in the case of mixed transitions.

Although different ways of exciting resonance fluorescence have been proposed[1] and more can be imagined, so far only radioactive isotopes decaying to the element under investigation have been successfully used as the source of the exciting radiation. The nuclear experiment is thus very similar to the resonance experiments with atoms,[2] in which, e.g., the light from a mercury arc is used to excite the resonance radiation in mercury vapor. The main difficulty in the nuclear case stems from the fact that for gamma rays the natural line width is, in general, small compared with the energy lost to the recoiling nuclei. Consequently, the exciting line is off resonance, the cross section is greatly reduced, and the resonance effect lost in the background of elastically scattered gamma rays. In order to observe nuclear resonance fluorescence, ways and means to restore the resonance condition have to be found. Up to the present, the Doppler shift due to mechanical motion[3] and the Doppler broadening due to thermal agitation[4] have been successful in producing observable nuclear resonance fluorescence effects. Both methods are, however, restricted to heavy nuclei and to gamma rays of less than about 500 kev, the limitations being imposed by the strengths of available materials and by the range of feasible temperatures.

Several authors[5,6] have suggested that the large velocities, imparted to the nuclei by the radiation preceding the gamma ray under study, could be utilized to restore the resonance condition. However, experiments on Mg^{24},[6] Fe^{56},[6,7] Cu^{63},[7] Mo^{96},[8] and Mo^{97},[9] using solid sources of Na^{24}, Mn^{56}, Zn^{63}, Tc^{96}, and Nb^{97}, were unsuccessful, although in these cases the preceding radiation has enough momentum to fully compensate for the recoil energy loss. The failure to observe resonance fluorescence in the above-mentioned isotopes can be attributed to the slowing down of the recoiling nuclei by collisions suffered prior to the gamma emission. The collision times encountered lie between 10^{-13} and 10^{-15} second. The absence of a resonance effect therefore establishes a lower limit of 10^{-12} to 10^{-13} second for the lifetimes of these gamma-ray transitions. In some favorable cases the increase in the slowing down time, achieved by the use of a liquid source, can be sufficient to make the resonance fluorescence effect observable.[7] However, our scanty knowledge of the slowing down process makes the evaluation of such experiments rather uncertain.

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

[1] L. I. Schiff, Phys. Rev. **70**, 761 (1946); A. M. Cormack, Phys. Rev. **96**, 716 (1954).

[2] See, e.g., A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, New York, 1934).

[3] 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).

[4] K. G. Malmfors, Arkiv Fysik **6**, 49 (1952); F. R. Metzger and W. B. Todd, Phys. Rev. **94**, 794 (1954); **95**, 627 and 853 (1954); F. R. Metzger, Phys. Rev. **97**, 1258 (1955) and **98**, 200 (1955).

[5] W. Kuhn, Phil. Mag. **8**, 625 (1929).

[6] E. Pollard and D. E. Alburger, Phys. Rev. **74**, 926 (1948).

[7] K. Ilakovac, Proc. Phys. Soc. (London) **A67**, 601 (1954).

[8] F. R. Metzger, Phys. Rev. **83**, 842 (1951).

[9] F. R. Metzger and W. B. Todd, Phys. Rev. **91**, 1286 (1953).

The obvious remedy to the difficulties stemming from the short collision times is the use of gaseous sources. Preliminary experiments with gaseous sources of Hg^{203} [10] and of Tl^{202} [11] were encouraging and it was decided to apply the method to an element among the lighter nuclei, i.e., in a region of the periodic table where the other methods of compensating for the gamma recoil energy loss are not applicable. Germanium was chosen for the first attempt because As^{74}, decaying to Ge^{74}, is available with high specific activity, has a relatively simple disintegration scheme and decays to the most abundant (36.7%) isotope of germanium.

The first experiments,[12] using sources of As^{74} vapor, demonstrated an easily observable resonance fluorescence effect approximately thirty times larger than the total elastic scattering. Subsequent runs with the shorter lived As^{72} showed effects of comparable magnitude in Ge^{72}. In both cases the angular distribution of the resonance radiation was measured with As vapor sources which, under the conditions then used, consisted almost exclusively of As_4 molecules.[13] For angular distribution studies the use of such sources is not objectionable; for the determination of the lifetimes, however, the exact shape of the exciting gamma line has to be known and the use of molecules with constituents of comparable mass makes the recoil phenomena, which are responsible for the line shape, rather complicated. For the lifetime measurements the use of As_4 sources was, therefore, abandoned and the experiments were carried out with AsH_3 sources.

MOMENTUM REQUIREMENT FOR THE PRECEDING RADIATION

The preceding radiation, through Doppler effect, has to supply the energy lost to the recoiling nuclei in the gamma emission and in the scattering process. For a gamma ray of energy E_γ, the recoil loss in the emission process is $E_R=E_\gamma^2/2Mc^2$, where M is the mass of the recoiling nucleus. The energy lost in the scattering process is the same, the total energy deficit ΔE is therefore given by $\Delta E=E_\gamma^2/Mc^2$. In order to restore the resonance condition, at least for those gamma rays emitted in the forward direction, the Doppler shift resulting from the preceding radiation has to equal ΔE, i.e., $(v/c)E_\gamma=E_\gamma^2/Mc^2$ or $P_{\text{prec}}=P_\gamma$. This means that, to be effective in restoring the resonance condition, the preceding radiation has to possess a momentum at least equal to the momentum of the gamma ray under study.

If the preceding radiation is itself a gamma ray or if it is a neutrino accompanying K-capture, the most favorable case is one in which the available decay energy is approximately equal to the energy of the radiation being studied. If the available decay energy is much larger than the necessary minimum, the magnitude of the resulting resonance effect is reduced accordingly.

For a beta decay with a maximum momentum just fulfilling the minimum condition, i.e., for a decay for which $P_{\max}=P_\gamma$, the observed resonance effect is small, because the number of beta decays giving the maximum amount of momentum is small. The most favorable decay energy for a beta decay as the preceding radiation is the one for which the maximum momentum is approximately twice the momentum P_γ.

The Doppler broadened line is centered around $E_\gamma=E_0-E_R=E_0-E_0^2/2Mc^2$, where E_0 is the excitation energy of the gamma emitting nuclear level. Exact resonance will occur for gamma rays of energy $E_{\text{res}}=E_0+E_R=E_0+E_0^2/2Mc^2$.

ANGULAR DISTRIBUTION OF THE RESONANCE RADIATION

(a) General

When a nucleus with a ground state of total angular momentum J_0 is excited with an unpolarized gamma-ray beam to a state with total angular momentum J, the angular distribution of the resonance radiation with respect to the exciting beam is identical with the angular correlation of successive gamma rays for the spin sequence J_0-J-J_0.[14] If the spin of the ground state is known, the determination of J is straightforward and involves fewer parameters than an angular correlation experiment, which necessarily involves two different gamma-ray transitions.

The observation of a sizeable resonance fluorescence effect implies that the lifetime of the intermediate state is shorter than 10^{-10} second and makes the influence of extranuclear fields on the angular distribution very unlikely.

Compared to the angular correlation experiment, the study of the angular distribution of the resonance radiation has the disadvantage of being, at least in most cases, restricted to angles between 90 and 150 degrees. The angles below 90 degrees are unfavorable because of the sharp increase of the elastic scattering and because of the difficulties arising from the large intensity of the Compton radiation. The necessity of shielding the detector from the direct beam by an absorber attenuating the direct radiation by a factor of the order of 10^6 imposes geometrical restrictions which render the angles beyond 150 degrees impractical. On the other hand, the symmetrical correlations of the type J_0-J-J_0, which are typical for the resonance radiation, are usually very large and the available range of angles is sufficient to determine the pertinent constants with the necessary accuracy.

[10] F. R. Metzger, Proceedings of the 1954 Glasgow Conference (Pergamon Press, London and New York, 1955), p. 201.
[11] F. R. Metzger, Phys. Rev. 98, 200 (1955).
[12] Reported at the Washington Meeting of the American Physical Society, April, 1955 [F. Metzger, Phys. Rev. 99, 613(A) (1955)].
[13] G. Preuner and I. Brockmöller, Z. physik. Chem. A81, 129 (1913).
[14] D. R. Hamilton, Phys. Rev. 58, 122 (1940).

(b) Experimental Procedure

The geometry used for the determination of the 120° point of the angular distribution of the 596-kev radiation in Ge^{74} is shown in Fig. 1. For the higher energy quanta of Ge^{72} a larger absorber of the direct beam was necessary and the geometry had to be modified accordingly.

The scattered radiation was detected with a thallium-activated sodium iodide crystal of 35-mm diameter and 40-mm length connected to a RCA 6342 photomultiplier tube and a single channel scintillation spectrometer. For the measurements of the differential cross sections the single channel accepted approximately the full width at half-maximum of the photopeak of the gamma ray under study. In order to prevent serious pile-up due to the pulses from the very intense Compton scattered radiation, a Cd-Pb shield surrounded the detector.

For all experiments the same ring scatterers were used. The germanium scatterer consisted of 25 bars of germanium metal[15] arranged in a ring of 11 inches diameter. Each bar, of approximately semicircular cross section, was $2\frac{1}{8}$ inches long and had a cross sectional area of $\frac{3}{4}$ square inches. For comparison purposes, a zinc ring scatterer of similar dimensions was fabricated from zinc sheet metal.[16] The scatterers were supported by three $\frac{3}{8}$-inch diameter aluminum rods. The different scattering angles were realized by changing the relative positions of source, scatterer and detector. Whenever possible only the detector was moved; in this way the solid angle subtended by the scatterer at the position of the source, which had to be calculated for the oddly shaped germanium bars, was kept constant. The change in the scatterer-detector solid angle, including the change in the absorption by the Cd-Pb shield, was determined by moving a point source to the different positions of the scatterer and by observing the resulting changes in the counting rates.

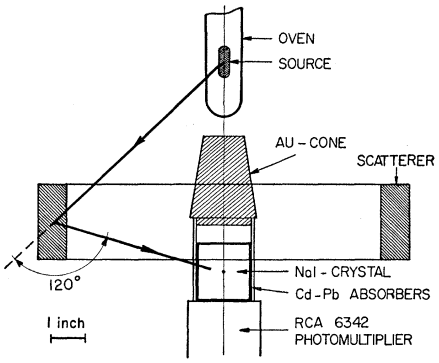

FIG. 1. Arrangement for Ge^{74} experiment. The relative position of source, scatterer and detector depicted is the one corresponding to a mean scattering angle of 120°.

[15] We are indebted to the Bell Telephone Laboratories, Murray Hill, New Jersey, for the loan of these germanium bars.

[16] Courtesy of The New Jersey Zinc Sales Company, New York, New York.

The As^{72} and As^{74} activities were produced by deuteron bombardment of germanium metal in the M.I.T. cyclotron.[17] The arsenic was separated from the germanium target by the method of Green and Kafalas.[18] The arsenic, now in acid solution, was reduced to arsine with zinc and the arsine was thermally decomposed to arsenic metal. 1.5 mg of arsenic was added to assure complete recovery of the active arsenic. However, in subsequent separations as little as 50 μg of As carrier was found to be ample. The arsenic metal was then distilled into quartz ampoules of volume 0.6 ml. The quartz ampoules, which had to be heated to 700°C in order to volatilize all the arsenic, were incorporated into air-tight stainless steel containers in order to prevent any contamination of the laboratory in case of rupture.

The oven consisted of 1-inch diameter quartz tubing wound with nichrome wire. As an additional safety measure, the quartz tubing of the oven formed a vacuum-tight vessel, which was evacuated and sealed off after the source in the stainless steel container had been inserted. The temperature of the source was measured with a chromel *P*-alumel thermocouple in contact with the stainless steel source container.

At a given angle the following procedure was adopted: First, with the source at several feet distance from the detector, the counting rate in the fixed channel was measured; this counting rate was taken to be representative of the source strength. The second step, with the source at room temperature, and in its normal position, consisted in a comparison of the counting rates due to the germanium and the zinc scatterers. Every five minutes the scatterers were exchanged and every twenty minutes the calibration of the scintillation counter was checked. For all positions the counting rates of the two scatterers agreed to better than 3% of the total rates, the total rates being of the order of 100 counts per minute for a source of about one millicurie.

After the comparison at room temperature the source was heated to 700°C. After temperature equilibrium

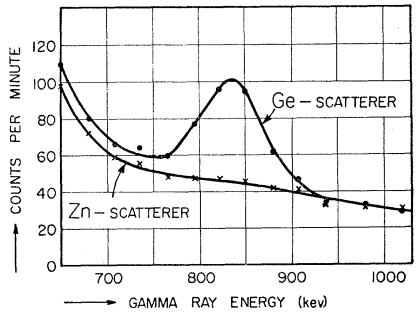

FIG. 2. Resonance fluorescence in Ge^{72}. Pulse-height distributions of the radiation scattered from zinc and germanium rings.

[17] H. J. Watters and J. F. Fagen, Phys. Rev. **92**, 1248 (1953).

[18] M. Green and J. A. Kafalas, J. Chem. Phys. **22**, 760 (1954).

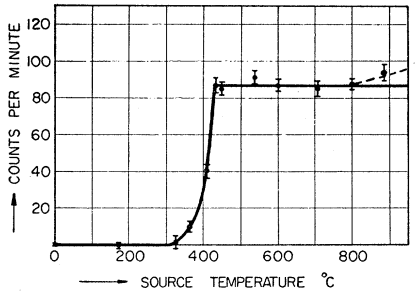

FIG. 3. Resonance fluorescence in Ge^{74}. Dependence of the resonance effect on the temperature of the arsenic source. The solid line represents the temperature dependence of the density of the As_4-vapor calculated on the basis of vapor pressure data,[13] and normalized to give the experimental value of the plateau. Above 800°C the dissociation of the As_4 vapor causes an increase (dashed line) of the resonance fluorescence effect (see discussion in the last chapter).

had been reached, the counting rate due to the germanium scatterer was compared with that due to the zinc scatterer using the same schedule as described above. While with the source at 700°C the counting rate with the zinc scatterer was practically unchanged, the counting rate with the germanium scatterer showed a considerable increase due to resonance fluorescence. For the source mentioned above the increase in the counting rate varied between 50 and 100 counts per minute for the different scattering angles.

In order to ascertain that the increase in counting rate, observed with the germanium scatterer, was due to resonance fluorescence from the 596-kev level in the case of the As^{74} source and from the 835-kev level in the case of the As^{72} source, the pulse-height distribution of the scattered radiation was measured for the two sources. Such a measurement was especially necessary in the case of As^{72}, because the decay scheme is uncertain and gamma rays with energies greater than 1 Mev are emitted. In Fig. 2 are reproduced the pulse-height distributions obtained with an As^{72} source for the germanium and the zinc scatterers. In the pulse-height distribution due to the radiation scattered from germanium, a clear photopeak corresponding to an 835-kev gamma ray is present. There is no indication of tails due to resonance fluorescence of higher energy gamma radiation. Further confirmation of this fact was derived from the observation that the germanium and the zinc scatterers gave rise to essentially identical counting rates in the region above 1 Mev. By an analogous experiment with As^{74} it was ascertained that in this case it is indeed the 596-kev radiation that gives rise to the increased counting rate when the germanium scatterer is in position.

The gaseous form of the arsenic sources is an essential condition for the observation of resonance fluorescence in Ge^{74}. This can be demonstrated by a measurement of the temperature dependence of the resonance fluorescence effect The results of an experiment with an ampoule containing 0.05 mg of As per ml are summarized

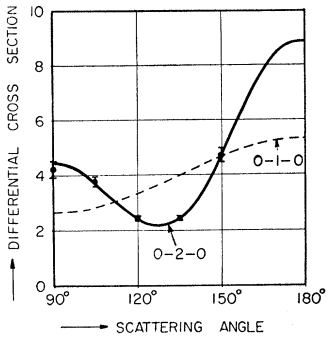

FIG. 4. Angular distribution of the resonance radiation from the 596-kev level in Ge^{74}. The solid line represents the theoretical angular distribution for an excited state with spin 2 and a ground state with spin 0, corrected for the finite angular resolution. The differential cross section is given in arbitrary units.

in Fig. 3. The resonance fluorescence effect follows the curve representing the number of arsenic molecules per ml of source volume calculated from the temperature of the source using known vapor pressure data.[13] Above 432°C, i.e., above the temperature at which all the arsenic in the ampoule is in the vapor phase, the effect is essentially constant. This latter fact is especially welcome for the angular distribution experiments because it disposes of the necessity of rigid controls for the source temperature.

Another indication that the thermal Doppler-broadening alone, even at 1000°C, is insufficient to compensate for the gamma recoil energy loss was obtained when a quartz ampoule broke inside the steel container. The resonance fluorescence effect suddenly disappeared, i.e., became smaller than 2% of the original effect, although all the activity was still in the steel container and the steel container was at 1000°C. The arsenic had chemically united with the steel in such a way that the compound formed was solid at 1000°C and thus did not give rise to resonance fluorescence.

(c) Results and Discussion

Germanium-74

The results obtained for the angular distribution of the resonance radiation from the 596-kev level in Ge^{74} are summarized in Fig. 4. The agreement of the experimental points with the theoretical distribution for an excited state with spin 2 and a ground state with spin[19] 0 is good. The least-squares fit, $W(\theta)=1+(0.37\pm0.07)P_2(\cos\theta)+(1.06\pm0.09)P_4(\cos\theta)$, has to be compared with the theoretical distribution, corrected for the finite angular resolution:

$$W(\theta)=1+0.37P_2(\cos\theta)+1.03P_4(\cos\theta).$$

It should be mentioned that for an even-A nucleus, as long as one considers only dipole and quadrupole transitions, the theoretical value $A_4/A_0=1.143$ for the 0–2–0 spin sequence is larger by more than a factor of 2 than the value A_4/A_0 for any other sequence J_0-J-J_0 with any mixture of dipole and quadrupole

[19] Townes, Mays, and Dailey, Phys. Rev. **76**, 700 (1949).

TABLE I. Observed and predicted ratios of the differential cross sections at 90° and 121°.

Spin of the 835-kev level	Theoretical ratio $d\sigma(90°)/d\sigma(120°)$	Experimental ratio
1	0.79	
2	2.04	2.2±0.3
3	0.08	

radiation. Therefore as soon as one is able to establish that A_4/A_0 is >0.51, the spin sequence involved must be 0–2–0, i.e., the spin of the ground state must be 0 and the spin of the excited state must be 2. The value $A_4/A_0=1.06\pm0.09$ which we measured for the 596-kev radiation of Ge^{74} therefore confirms the assignment[19] of spin zero to the ground state of Ge^{74}.

Germanium-72

Because of the relatively short half-life of As^{72} (26 hours) the angular distribution measurements with the 835-kev radiation were restricted to two angles, 90° and 121°. The ratio of the differential cross sections for resonance fluorescence at these two angles was found to be 2.2±0.3. For Ge^{72}, with spin zero in the ground state,[19] this ratio of differential cross sections depends only on the value of the spin of the 835-kev excited state. The values of the ratio, expected for different spins of the 835-kev state, are given in Table I. From this tabulation, one concludes immediately that the spin of the 835-kev level in Ge^{72} is 2. This value of the spin, and the fact that Coulomb excitation of this level has been observed,[20] make the parity of the 835-kev state even and characterize the 835-kev transition as an electric quadrupole. It should be mentioned that the 835-kev state is not the first excited state of Ge^{72}; in violation of the Goldhaber-Sunyar[21] rule, the first excited state of Ge^{72} is a 0^+ state[22] at ~700 kev.

DETERMINATION OF THE GAMMA-RAY LIFETIMES

(a) Absolute Cross Section and Lifetime

The mean life of a gamma-ray transition to the ground state of a nucleus is closely related to the cross section for resonance fluorescence. If gamma emission to the ground state is the only mode of decay for the excited state, i.e., if internal conversion is negligible and if no branching exists, a detailed balance consideration[2] shows that

$$\int_{\text{line}} \sigma(E)dE=(g_2/g_1)\cdot\frac{\lambda_0^2\hbar}{4\tau},$$

where λ_0 is the wavelength at resonance, τ is the mean life of the level, g_1 and g_2 are the statistical weights of the ground state and the excited state, respectively, and $\sigma(E)$ is the cross section for resonance fluorescence for a gamma ray of energy E.

Because of the recoil from the preceding radiation, the exciting gamma radiation has a breadth that is so wide in comparison with the absorption line width that it can be regarded as a continuous spectrum. The energy distribution $N(E)$ in this spectrum can be calculated from the properties of the preceding radiation which are usually known. The cross section for resonance fluorescence, averaged over the incident spectrum, is $\sigma_{Av}=\int N(E)\sigma(E)dE/\int N(E)dE$. In view of the sharpness of the absorption line, $N(E)$ can be taken as being constant in the region where σ is significantly different from zero, i.e., in the neighborhood of the resonance energy E_{res}. We then have

$$\sigma_{Av}=\frac{N(E_{res})\int\sigma(E)dE}{N}=\frac{N(E_{res})}{N}\frac{g_2}{g_1}\frac{\lambda_0^2\hbar}{4\tau}.$$

Solving for τ, expressing λ_0 in terms of E_{res}, and using the numerical values for the constants, one obtains

$$(\tau)_{sec}=\frac{g_2}{g_1}\frac{2.53}{E_{res}^2\sigma_{Av}}\frac{N(E_{res})}{N}, \quad (1)$$

where E_{res} is measured in ev, σ_{Av} in barns and where $N(E_{res})$ is the number of gamma rays in a one-ev internal at $E_{res}=E_0+E_0^2/2Mc^2$.

Knowing already the angular distribution, the main problem consists in finding a source for which $N(E_{res})$ can be calculated and in then determining σ_{Av} for this source by a measurement of the differential cross section at one angle.

(b) Line Shape of the Exciting Radiation, Calculation of $N(E_{res})/N$

The final values for the lifetimes of the 596-kev transition in Ge^{74} and the 835-kev transition in Ge^{72} were obtained from experiments with AsH_3 sources; we shall therefore discuss in some detail the calculations of the line shapes for AsH_3 sources.

As far as the recoil behavior is concerned, AsH_3 (arsine) is the closest approximation to monatomic arsenic vapor that can be prepared without much difficulty. Because of the large ratio of the masses of the constituents of the arsine molecule, the energies available for excitation of the molecule when the As atom receives its recoil are small[23] and are not sufficient to break up the molecule. One therefore expects the arsine molecule to recoil as a whole and one consequently carries out the calculations of the line shape using the molecular weights 77 and 75 for $As^{74}H_3$ and $As^{72}H_3$, respectively. The disintegration schemes of the two isotopes are very different and the two cases have to be treated separately.

[20] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **99**, 617(A) (1955).
[21] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[22] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).
[23] Hans Suess, Z. physik. Chem. **B45**, 312 (1940).

Arsenic-74

According to the decay scheme[24] of As^{74}, 60% of the disintegrations leading to the 596-kev level in Ge^{74} proceed by electron capture, 40% by positron emission. An analysis of the pulse-height distribution due to the As^{74} gamma rays, observed with a scintillation spectrometer, leads to a value of $59\pm4\%$ for the share of the electron capture transitions, and of $41\pm4\%$ for the positron decays, in good agreement with the accepted disintegration scheme.

The energy available for the K-capture transition to the 596-kev state in Ge^{74} is 1.94 Mev, the momentum of the neutrino is 3.78 mc. The molecules containing decaying As^{74} nuclei acquire a recoil momentum equal in magnitude and opposite in direction to that of the neutrino. Depending on the angle of emission with respect to the recoil motion, the gamma rays exhibit Doppler shifts ranging from -16.06 ev to $+16.06$ ev. As long as the neutrono angular momentum is one-half, the gamma emission is isotropic with respect to the recoil direction and all the Doppler shifts in the interval given above occur with equal probability. The line shape from K-capture alone is thus a rectangle of 32.1-ev width, centered around $E_\gamma=E_{\text{res}}-\Delta E$, where $\Delta E=E_\gamma^2/Mc^2=5.15$ ev. The superposition of the thermal Doppler broadening and of the Doppler broadening due to the x-rays and the Auger electrons results in a rounding off of the corners of the rectangular distribution, but fails to affect the region near E_{res}.

While the recoil momentum spectrum for the K-capture transition consists of a single line, the recoil distribution due to the 0.92-Mev positron spectrum is continuous, ranging from zero to the maximum recoil momentum of 2.62 mc. The maximum Doppler shift corresponding to this momentum is 11.1 ev. The gamma line caused by the positron decay is therefore 22.2 volts wide at its base; it is symmetrical with respect to $E_\gamma=E_{\text{res}}-5.15$ ev and its exact shape depends on the beta-neutrino and the recoil-gamma angular correlations. To simplify matters, isotropic beta-neutrino and beta-gamma angular correlations were assumed. Calculations with beta-neutrino correlations of the form $[1\pm(p_e/E_e)\cos\theta]$ showed that, for the particular case of Ge^{74}, the fraction of gamma rays per ev interval at E_{res}, $N(E_{\text{res}})/N$, which essentially determines the magnitude of the resonance effect, is practically independent of the assumed beta-neutrino correlation. The values of $N(E_{\text{res}})/N$ for the strong correlations indicated above differ from the value $N(E_{\text{res}})/N$ for the isotropic correlation by less than 4%.

Using a ratio of 59 to 41 for the contributions of K-capture and of positron decay, one obtains the composite gamma line shape depicted in Fig. 5. From this graph the fraction of quanta per ev interval at E_{res}, $N(E_{\text{res}})/N$ is found to be 4.00%. If one uses the limiting values for the ratio K/β^+, i.e., 1.22 and 1.70,

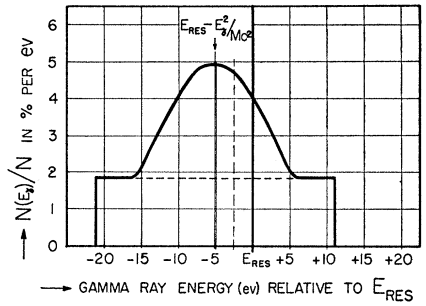

FIG. 5. Line shape of the 596-kev gamma rays emitted by an AsH_3 source, calculated on the basis of 59% electron capture and 41% positron decay. The rounding-off effects of the thermal motion and of x-rays, etc. are neglected. The dashed vertical line indicates the position of E_0, the energy of the excited state of Ge^{74}.

$N(E_{\text{res}})/N$ varies only from 4.09 to 3.92. Thus the uncertainty in our knowledge of the branching ratio contributes little to the error in the value for the lifetime.

In view of the insensitivity of $N(E_{\text{res}})/N$ to uncertainties in the decay scheme and in the beta-neutrino angular correlation, the error in $N(E_{\text{res}})/N$ was estimated to be smaller than 5%, i.e., $N(E_{\text{res}})/N=(4.00\pm0.2)\%$ per ev.

Arsenic-72

For this isotope the uncertainties in the disintegration scheme are considerable. It is known[25] that 62% of the positron decays lead directly to the 835-kev level in Ge^{72}, that 19% go to the ground state while the remaining 19% feed different levels above the 835-kev state. The amount of electron capture, however, is uncertain. McCown, Woodward, and Pool[26] report x-rays and positrons in a ratio of 2 to 1. A study of the relative intensity of annihilation radiation and 835-kev gamma rays leads to a positron to 835-kev gamma-ray ratio of 0.73 ± 0.20. Using allowed capture-positron branching ratios[27] we have calculated, on the basis of the reported intensities of the partial positron spectra, the electron capture intensities feeding the four excited states at 0.835-, 1.50-, 2.67- and 3.07-Mev excitation energy. In order to bring the positron to 835-kev gamma ratio into accord with the experimental value, the intensity of the 0.27-Mev positron spectrum had to be reduced. Assuming that each of the higher levels reaches the 835-kev level by a single gamma ray, the values $N(E_{\text{res}})/N$ were then calculated for the four different decay paths. The results of these calculations are summarized in Table II.

In view of the considerable uncertainties pointed out above, the error in the final value for $N(E_{\text{res}})/N$ has been estimated at $\pm20\%$, i.e., $N(E_{\text{res}})/N=(1.98\pm.4)\%$ per ev.

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[25] Mei, Mitchell, and Huddelston, Phys. Rev. **79**, 19 (1950).

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(c) Measurement of σ_{Av}

Germanium-74

The geometry used for the absolute measurement of the differential cross section was essentially that of Fig. 1. The use of a source ampoule of different size changed the effective angle to 118°.

The source consisted of 50 micrograms of arsine contained in a Pyrex ampoule of 1.7 ml volume. At room temperature this amount of arsine will produce a pressure of approximately 7 mm Hg, i.e., a pressure low enough to eliminate collision effects.

Although the comparison of the zinc and germanium scatterers during the angular distribution experiments indicated that, in the absence of resonance fluorescence, the two scatterers were well matched, another comparison with the arsine source at liquid air temperature was carried out. The difference in this case amounted to $+0.2\pm2.3$ counts per minute; the plus sign indicates that the germanium showed the slightly higher counting rate. The total counting rate was 110 counts per minute. After removing the liquid air, i.e., with the source again at room temperature and in the gaseous phase, the counting rate with the germanium scatterer exceeded that with the zinc scatterer by 179.4 counts per minute. The resonance effect thus amounted to 179.2 counts per minute.

Several similar runs were carried out. In between runs the distribution of the As^{74} activity over the source volume was measured by observing, through a lead slit of $\frac{1}{16}$ inch width and 4 inches thickness, the counting rates due to different portions of the ampoule. These measurements indicated a slow decomposition of the arsine gas which had to be taken into account.

Using the calculated value for the solid angle source-scatterer and the experimentally determined solid angle scatterer-detector, and taking into account the results of the angular distribution experiments, one obtains from all runs a value $(7.5\pm0.2)\times10^{-26}$ cm^2 for σ_{Av} of Ge^{74} for the 596-kev radiation emitted by an arsine source at room temperature. To arrive at this value, the finite thickness of the scatterer and the preferred attenuation of the resonance radiation in the scatterer had to be taken into account and the source strength measurements had to be corrected for the contributions of the annihilation radiation and of the 635-kev gamma ray to the counting rate in the fixed channel.

TABLE II. Relative intensities of the different branches of the As^{72} decay and corresponding values of $N(E_{res})/N$.

Level excitation energy, Mev	Intensity of positrons	Intensity of K-capture	$N(E_{res})/N$ percent per ev	Average $N(E_{res})/N$
0.835	63	3	1.99	
1.50	12	1	2.33	1.98
2.67	5	13	1.91	
3.07	<1	40	1.88	

Germanium-72

For the 835-kev transition in Ge^{72}, the measurement necessary for the determination of σ_{Av} was carried out at an effective angle of 90 degrees. As in the case of Ge^{74}, the source consisted of arsine gas at one hundredth of an atmosphere pressure.

Without resonance fluorescence, i.e., with the source at liquid air temperature, the two scatterers gave rise to practically identical counting rates, the difference being -0.2 ± 2.8 counts per minute. The total counting rate with the zinc scatterer was 76 counts per minute. With the source at room temperature the counting rate with the germanium scatterer was by 63.7 counts per minute larger than the counting rate with the matched zinc scatterer. The resonance fluorescence effect thus accounted for 63.9 counts per minute. After the necessary corrections, which were similar to those made in the case of Ge^{74}, a value $(7.8\pm.3)\times10^{-26}$ cm^2 was obtained for σ_{Av} of Ge^{72} for the 835-kev radiation emitted by an arsine source at room temperature.

(d) The Mean Lives of the Gamma Transitions

In Table III are summarized the numerical values of the different terms which enter into the calculation of the mean lives according to formula (1).

The errors of the σ_{Av} values are no longer the statistical uncertainties, but represent the estimated combined uncertainties in the source strength determination, the solid angle calculations, the absorption corrections, etc.

Recently,[20] Coulomb excitation of the two levels has been reported. The mean lives calculated from thick-target yields[28] are 1.3×10^{-11} second for the 596-kev transition in Ge^{74} and 1.9×10^{-12} second for the 835-kev transition in Ge^{72}. The agreement in the case of the 596-kev transition is satisfactory, the seriousness of the discrepancy in the mean lives of the 835-kev transition depends on the uncertainty attached to the mean life deduced from the Coulomb excitation yield measurement. Comparisons in other cases will indicate whether there exists a definite trend in the discrepancies between the lifetimes measured with these two methods.

The mean lives calculated on the basis of the single-particle model[29] are 2.5×10^{-10} sec and 4.7×10^{-11} sec, i.e., they are 13 and 10 times longer than the experimental mean lives. This indicates that even in these relatively light nuclei cooperative phenomena are quite pronounced.

INFLUENCE OF MOLECULAR BINDING AND OF COLLISIONS

Only a few elements are available as monoatomic vapors at reasonable temperatures, i.e., below the softening point of quartz. If one wishes to apply the resonance fluorescence method to other elements, volatile compounds will have to be used. This im-

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TABLE III. Mean lives of the two gamma-ray transitions and experimental data used to calculate them.

E_{res} (ev)	$(E_{res})^2$	$\frac{g_2}{g_1}=\frac{2J_e+1}{2J_g+1}$	σ_{Av} (barns)	$N(E_{res})/N$	τ_{sec}
5.96×10^5	3.55×10^{11}	5	$(7.5\pm1.1)\times10^{-2}$	4.00 ± 0.2	$(1.9\pm0.3)\times10^{-11}$
8.35×10^5	6.97×10^{11}	5	$(7.8\pm1.2)\times10^{-2}$	1.98 ± 0.4	$(4.6\pm1.2)\times10^{-12}$

mediately introduces the problem of the sharing of the recoil energy among the constituents of the molecule and of the breakup of the molecules.

Some indication of these difficulties appeared when the absolute cross sections for resonance fluorescence in Ge^{74}, measured with As_4 and with AsH_3 sources, were compared. The σ_{Av} determined in the angular distribution experiment with As_4 sources was smaller than the σ_{Av} measured with AsH_3 sources, the ratio of the two cross sections being 0.76. As both measurements must lead to the same value for the mean life of the 596-kev transition, the expressions $N(E_{res})/N$ for the two sources have to be in the same ratio of 0.76 as the σ_{Av}'s [see Eq. (1)]. If one assumes a breakup of the As_4 molecule to occur whenever the recoil energy of the Ge* nucleus exceeds the binding energy of an As atom to the As_4 molecule, one arrives at a value of 0.92 for the ratio of the values of $N(E_{res})/N$ for As_4 and for AsH_3 sources. The discrepancy between this value and the experimental value 0.76 was at first attributed to the considerably higher vapor pressures in the As_4-sources used for this comparison, i.e., the difference was attributed to changes in the line shape due to collisions. However, when experiments with As_4 sources differing by as much as a factor of 1000 in vapor density were carried out, they yielded the results summarized in Fig. 6. These results show that, due to a favorable mixture of positron decay and K-capture, the resonance fluorescence effect in Ge^{74} is very insensitive to collisions in the As_4 sources. Evidently the loss in $N(E_{res})/N$ due to the collision-narrowing of the β^+ portion of the gamma line is compensated by a gain in $N(E_{res})/N$ due to the contraction and consequent increase in height of the K-capture rectangle (see Fig. 5). A calculation shows that the first four collisions between Ge* nuclei and As_4 molecules change $N(E_{res})/N$ by less than 10%; the first collisions actually cause a slight increase of $N(E_{res})/N$. Using a molecular collision diameter of 4.6×10^{-8} cm the solid curve in Fig. 6 was calculated. It indicates that, once the As_4 density is below $\sim$15 mg/ml, $N(E_{res})/N$ varies by less than 3%.

In AsH_3 sources the collisions take place between particles of equal mass, the resonance fluorescence effect excited in Ge^{74} by the radiation from AsH_3 sources is therefore more sensitive to collisions. However, for the very low pressures used for the measurement of σ_{Av} the influence of collisions is smaller than 1%.

Having shown that the collisions between the recoiling Ge* nuclei and As_4 molecules are not responsible for the low value of $N(E_{res})/N$ deduced from the measurements with As_4 sources, another explanation had to be found.

Realizing that $N(E_{res})/N$ is zero for decays which do not break up the As_4 molecules, one is led to assume that 17% of the expected breakups did not occur or did so only after the momentum of the Ge* nuclei had been distributed among the constituents of the molecule. Presumably these 17% of the decays are those in which the Ge* nucleus has a kinetic energy larger than the bond energy but moves towards the triangle formed by the other three atoms of the original As_4 molecule, i.e., they are decays in which the bonds are compressed. Further experiments with different arsenic compounds or with one arsenic compound and different recoil conditions would help to clarify the situation.

As a consequence of the difference in the $N(E_{res})/N$ values for As_4 sources, and for sources of approximately free As atoms, one expects to observe a change in the resonance fluorescence effect from an arsenic source when one reaches temperatures at which dissociation into As_2 and As becomes appreciable. For the high-density sources used in the angular distribution experiment, no such effect was noticeable even at 1150°C. However, for the low-density source, used in obtaining the data presented in Fig. 3, a gradual increase of the resonance fluorescence begins at 800°C (dashed line in Fig. 3). The total increase between 800°C and 1150°C amounts to 16%, a value consistent with the dissociation data of arsenic vapor[13] and with the difference between σ_{Av} for As_4 and for AsH_3 mentioned earlier.

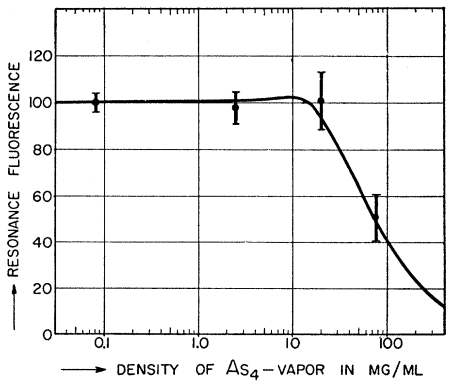

FIG. 6. Influence of collisions in As_4 sources on the resonance effect from the 596-kev level in Ge^{74}. The solid curve was calculated using a molecular diameter of 4.6×10^{-8} cm and an original line shape very similar to that given in Fig. 5.

In view of Ilahovac's[7] positive results with liquid sources of Zu^{63}, we looked for resonance fluorescence effects from the germanium scatterer using liquid sources of As^{72} and of As^{74}. In both cases the result was negative, i.e., the observed effect, if any, was smaller than 3% of the effect measured with a gaseous source of the same strength.

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Neutron Cross Sections for Zirconium

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The inelastic neutron cross section for the excitation of the 930-kev level in zirconium has been measured as a function of neutron energy between 0.9 and 2.2 Mev. The de-excitation gamma radiation has been observed with a single-crystal NaI(Tl) spectrometer. Isotopic assignment of the observed level is still uncertain. The total cross section for zirconium has been measured for neutrons with energies between 0.7 and 1.2 Mev, using a hydrogen recoil counter in a good-geometry transmission experiment. Some correlation between inelastic and total cross sections has been observed.

I. INTRODUCTION

THE inelastic scattering of neutrons by nuclei has been a subject for much experimentation during the last few years. Several different techniques for the determination of cross sections have been worked out, and are discussed in the literature.[1] Determinations of the inelastic cross section as a function of bombarding neutron energy have been made for a number of elements,[2,3] mainly those with first excited states of 800 kev or more. Resonance structure observed in the cross sections for Al, Cr, Ni, and Fe has been attributed to resonances in the formation of the compound nucleus. Griffith[4] has determined the inelastic cross section for Zr for 4.5-Mev neutrons. The purpose of the work reported here was to observe the inelastic cross section for zirconium as a function of incident neutron energy, and to endeavor to compare it with the total cross section, in both magnitude and resonance structure.

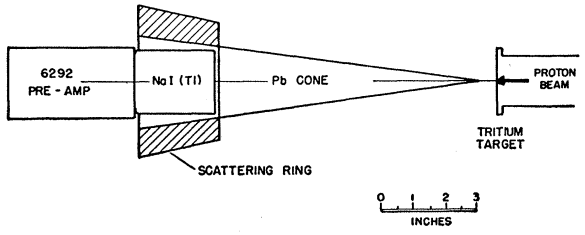

FIG. 1. Large-crystal experimental geometry. The single-crystal NaI(Tl) spectrometer is mounted inside, and coaxially with, the conical scatterer. The 8-inch lead cone effectively shields the crystal from neutrons and gamma rays coming from the tritium target. The resolution of neutrons striking the scattering cone is about 20 kev, the solid angle subtended by the scatterer at the crystal is close to 2π.

II. EXPERIMENTAL METHOD

Inelastic Cross Section

The method chosen for this investigation was the same as that described by Kiehn and Goodman.[2] A single-crystal NaI(Tl) spectrometer is used to observe the gamma-ray spectrum arising from the de-excitation of those levels in the scattering sample which have been excited by the inelastic scattering of the incident neutrons. The scatterer was made of natural zirconium, in the form of a hollow truncated cone (i.d.$=2\frac{1}{4}$ in., o.d.$=3\frac{3}{4}$ in., thickness$=2\frac{1}{2}$ in. at the face presented to the neutron beam). This was placed on the axis of symmetry of the incident neutron beam, as shown in Fig. 1. The detector was a 2 in.$\times$2 in. Harshaw

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