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1. INTRODUCTION

Atomic and molecular beams provide one of the oldest methods of experimental physics. Their earliest applications were, as to be expected, to the kinetic theory of gases, because study of the effusion of gas molecules from a source was of fundamental importance in understanding the Maxwellian theory of gases. The technique was soon recognized, however, as especially valuable for the study of molecular, atomic, and - later - nuclear structure. In a beam, the atomic or molecular system is in effect isolated from any other molecule or atom. Thus, in a study of atomic, molecular, or nuclear properties, all experimental complications arising from the effects of the solid, liquid, or gaseous state can be avoided. If the experiment utilizes a resonance method, the lower limit to the width of the resonance line depends only upon the radiation lifetime and the duration of observations.

Advantages of the reaction-free state become apparent from consideration of the numbers involved in a typical experiment with a nonradioactive

beam. Such a beam consists of 10^{10} or 10^{11} atoms per second at the detector in an apparatus whose length is of the order of 1 meter. The distribution of velocities is the Maxwellian distribution multiplied by the velocity, v. The mean velocity may be 50,000 cm/sec, giving a lineal density of $\sim 10^{6}$ atoms/cm; for a beam 0.01 cm wide and 1 cm high, this is a volume density of $\sim 10^{8}$ atoms/cm^3. The result is a much smaller collision rate than in a standard gas density of 10^{19} atoms/cm^3. Furthermore, if the cross section for collision of the beam atoms with themselves is spherically symmetric, the probability that products of collisions will remain in the beam is only a little greater than the apparatus transmission. Since this is about 10^{-5} or less, the effect of collisions is negligible. Scattering due to residual gas in the apparatus is scarcely significant: the pressure of the residual gas is 5×10^{-7} mm of Hg, or less, which corresponds to a mean free path ten times the length of the apparatus.

The molecular (or atomic) beam became an important device for modern physical research when O. Stern and his collaborators used it for studying such diverse phenomena as molecular scattering and space quantization.[1, 2, 3] In these experiments he studied the deflection of the atomic system in very strong inhomogeneous magnetic fields.[4, 5] The specific conformation of magnet pole tips required to produce this inhomogeneity still goes by the name Stern-Gerlach field.

The next major step in the technique, based on an interesting theoretical observation by Breit and Rabi,[6] was put into experimental practice by Rabi and his collaborators.[7, 8, 9] This concerned the

strong magnetic interaction between the currents produced by the electron angular momentum of the atom and the nuclear magnetic moment. The nuclear spin is rather strongly coupled to the electronic angular momentum, and the two systems would not be decoupled in a magnetic field of less than several hundred gauss.* In a weak field, therefore, the two systems behave

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*Throughout this paper all magnetic moments are in units of the Bohr magneton, μ_o, about 0.927×10^{-20} ergs/gauss. The vector magnetic moment of the electron system is $g_J \mu_o \underset{\sim}{J}$, where $\underset{\sim}{J}$ is the vector angular momentum in units of $\hbar$, and g_J is known as the gyromagnetic ratio of the system and depends upon the details of the electronic motion. For a single electron in an s state, we have $g_J \cong -2$, and, in general, $g_J \sim -1$. For a nucleus, however, the magnetic moment is $g_I \mu_o \underset{\sim}{I}$, where $\underset{\sim}{I}$ is the vector nuclear spin in units of $\hbar$, and, what is important, $g_I \sim 1/2000$. As an example, for a single s electron, the magnetic field at the nucleus due to the spin of the electron is roughly $2\mu_o/r^3$. If r is taken as $\approx 0.5 \times 10^{-8}$ cm, this is equivalent to about 10^5 gauss. A unit nuclear moment in this field of 10^5 gauss is therefore equivalent to about 5×10^{-19} ergs. When this is divided by Planck's constant, one gets 10^8 cycles/sec or 100 Mc/sec, if frequency is used as a unit of energy. This represents a rather strong coupling of the nuclear spin to the electronic angular momentum, and, since the electron precession frequency in a magnetic field, if uncoupled, is $g_J \mu_o H/h$, or about 1.4 Mc/sec, for $g_J = 1$, it would take a field of several hundred gauss to completely decouple the two systems.

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as one, with an angular momentum that is the vector sum of the two angular momenta, but with a magnetic moment that, to a very good approximation, is due to the electronic structure alone. Thus the behavior of the system as a whole in an external field is primarily influenced by the spin of the nucleus, and not by its magnetic moment, except through the hyperfine coupling. This was the basis for the zero-moment method used by Rabi and his collaborators for the measurement of the spins, magnetic moments, and quadrupole moments of the stable isotopes of the alkalis, gallium, and indium. (The Breit-Rabi diagram is later explained in some detail.) The next step in the development of the subject was the discovery and application by Rabi of the nuclear radiofrequency resonance method to molecular beams. This was an advance of many orders of magnitude in the precision of measurement of nuclear moments, and the application of the method to HD and D_2 led to the discovery and measurement of the deuteron quadrupole moment.[10] Immediately afterward, the method was applied to the radiofrequency spectroscopy of alkali atoms by Millman, Kusch, and Rabi,[11] and because of its enormous precision, virtually supplanted the zero-moment method. Zacharias measured the spin of K^{40}, the rare radioactive isotope of potassium, by an ingenious application of the method, and thereby demonstrated its applicability to trace amounts of material in the presence of large numbers of background atoms.[12] The resonance method was extended to measurements of quadrupole interaction in nuclei, including what is known as "pure quadrupole resonance".[13] The Kusch-Foley measurements established the anomalous magnetic moment of the electron[14] and set the stage for the rapid development of the renormalized-electron-field

theory and Lamb's experiment leading to the discovery of the "Lamb shift".[15]

For a broad discussion of procedures and techniques of atomic beams, the reader should refer to several articles that cover all the above applications.[16] This discussion is primarily concerned with the applications to the measurement of the spins, magnetic moments, and quadrupole moments of unstable nuclei of short half-lives. These quantities are of primary importance in the study of nuclear structure, but until recently there has been very little done in the field, considering the large number of known isotopes. As a result, although much has been known about the nuclei that lie along the "stable line" of the Segrè chart and long-lived isotopes that may be produced in abundance from these in reactors, very little was known about those isotopes that are neutron-deficient by two, three, four, or more neutrons. The primary reason is that these are cyclotron-produced isotopes and, in general, are not available in quantities greater than about 10^{13} atoms. By an amalgamation of the techniques of nuclear physics with atomic beams, measurements have been successfully carried out on 10^{10} atoms and on species whose half-lives are one-quarter hour. Within the last two years, the nuclear spins of about fifty isotopes have been determined by this extension of the method of atomic beams. The half-lives of these isotopes have ranged from 1/4 hour to 24,000 years. It should be emphasized that these measurements have been made with no loss of the great resolution available and thus have a very high reliability compared with other methods. The usefulness of the results can be appreciated from the fact that nuclear spins are now known for ten isotopes of Cs, ranging from a magic number of neutrons for Cs^{137} to ten less than the

magic number of neutrons for Cs^{127}. In addition, long-lived isomeric states have been investigated. Much useful information has been obtained on Au, and on Ag isotopes. Because these atoms are in an electronic state with $J = 1/2$, it is not feasible to measure the quadrupole moments of these nuclei. Instead, an attempt is being made to observe the hyperfine-structure anomaly for these nuclei. This term is also nuclear-shape-dependent. At present, work is proceeding on the halogen, Ga and In isotopes. The advantage to be gained is that measurements can be made on a $J = 3/2$ state, and a quadrupole (as well as an octupole) term can be measured. If this is possible, it will result in the knowledge of the quadrupole moments for eight or nine isotopes of the same element and should be extremely useful in the interpretation of the unified nuclear model and similar theories.

2. ISOTOPE PRODUCTION AND PREPARATION

In this section, the problem of producing sufficient isotope for detection and measurement is analyzed, and some numerical examples are given. To fix the ideas, the examples of Rb^{81}, Rb^{82}, Rb^{83}, and Rb^{84} are discussed. The detailed discussion of the atomic beam techniques and apparatus is reserved for the next section.

The stable isotopes of Rb are Rb^{85} and Rb^{87}, with spins of 5/2 and 3/2 respectively. Their nuclear magnetic moments are 1.35 and 2.75 in units of the nuclear magneton, 5.05×10^{-24} erg/gauss. The most convenient way to produce the neutron-deficient isotopes is via alpha bombardment of Br. Since Br has two stable isotopes in nearly equal abundance, Br^{79} and Br^{81}, there may be more than one way to produce the same isotope. If 50-Mev alpha particles are available, reactions of the type (α,kn) are

possible, where k = 1, 2, 3, or 4. Other reactions are possible, but the yield of radioisotope is so low that they may be neglected. Furthermore, we will see that the identification, by means of chemistry, vapor pressure, electronic structure, and characteristic x-rays, is sufficiently rigid to remove any possibility that the finally detected beam is not rubidium. Figure 1 is a production curve for alpha-particle reactions, estimated from the evaporation model. These curves and the initial energy of the alpha particles can be used in a numerical integration to estimate the yield of the radioisotopes. The numbers used as an example are based on an actual run at the Berkeley 60-inch cyclotron.

The overriding problem in working with the neutron-deficient isotopes of short half-life is one of intensity. It is important to obtain yields as great as possible from a given cyclotron bombardment. This means that the largest possible beam should be used. In working with isotopes of Au that can be made by alpha particles on Ir, this is no problem, because the heat developed on a conventional water-cooled target can never be enough to melt or vaporize the Ir or Au under reasonable circumstances (up to about 100 μa of beam at ~ 45 Mev). However, for most substances, such as the bromides in this example, a beam of only a few microamperes at this energy vaporizes and decomposes the salt in a conventional holder. Therefore, a special target assembly (Fig. 2) was designed to permit bombardment by currents up to 40 μa without destruction of the salt. For estimation of the number of atoms involved, reference is made to Fig. 1, which gives the differential cross section for each of the four reactions (α,kn) on bromine, for k = 1, 2, 3, 4. Knowing the density of $BaBr_2$ and

interpolating an appropriate range-energy relation for alpha particles, one can reach the estimates given in Table I. The yield for an average cyclotron beam current of 24 μa and of 45-Mev alpha particles is also given in the table, as well as the number of disintegrations per minute immediately after a 5.75-hr bombardment. These numbers can be most useful if they are translated to counts per minute. The counters used are K x-ray counters (more fully described later). We use here only those numbers necessary for intensity estimates. First, we have the factors common to all four isotopes:

(a) Counter solid-angle factor of 0.5.

(b) Window efficiency - the effect of the finite resolution of the counter discriminator. This is about 0.85.

(c) A fluorescent yield of 1/1.62.

(d) A factor of 10/11 for K versus L capture.

Combined, these factors give a counting efficiency of 0.24. Now there are factors that are different for each isotope:

Rb^{81}: 0.87 for the electron capture versus β^+ emission.

Rb^{82}: 0.94 for the electron capture versus β^+ emission. In addition, there is a guessed factor of 1/2 for competition in production with the metastable Rb^{82} that does not decay to the ground state.

Rb^{83}: The low-energy gamma rays of Kr^{81m}, the daughter, which augments the counting by a factor 1.4.

Rb^{84}: A factor of 0.76 because of electron branching.

The total predicted counting rate immediately after bombardment is

Table I. Results of Br (α,kn) Rb reactions from bombardment of Br with a cyclotron beam of 45-Mev alpha particles at 24 μa (average).

	Rb^{81}	Rb^{82}	Rb^{83}	Rb^{84}
Total effective production cross section ($cm^2 \times 10^{-25}$)	3.19	3.06	1.50	0.31[a]
Number of atoms produced per minute ($\times 10^{12}$)	1.0	0.97	0.47	0.1
Equivalent number of disintegrations per minute after 5.75-hr bombardment ($\times 10^9$)	570	420	0.94	0.49

[a]Reaction not completely realized because energy available (45 Mev) takes in only half the (α,4n) peak.

Rb^{81}: 1.2×10^{11} cpm

Rb^{82}: 0.5×10^{11} cpm

Rb^{83}: 3.0×10^{8} cpm

Rb^{84}: 0.9×10^{8} cpm

This can be compared with the results obtained from the analysis of a sample from the "straight-through" or undeflected beam. The ratio of the observed counting rates for the $Rb^{83} + Rb^{84}$ components to $Rb^{81} + Rb^{82}$ is 2.5×10^{-3} predicted and 1.9×10^{-3} observed. A rough absolute check also can be undertaken. The sample used for beam analysis was 0.02 of the total beam determined by monitoring the beam carrier. The transmission of the apparatus is $3.3 \times 10^{-5}/\pi$, and the decay rate (extrapolated) for the sample was 13,700 cpm, giving 0.7×10^{11} cpm total, or about 1/2 that predicted. This is very good agreement, considering the nature of the estimates. There is still another check on the intensities, and that involves the intensities of the observed resonances. The spins of Rb^{81}, Rb^{82}, Rb^{83}, and Rb^{84} are 3/2, 5, 5/2, and 2, respectively; their resonances were observed under many magnetic-field and production conditions. If each of the counting rates at the spin resonances is normalized to the particular component present in the beam and plotted versus $1/(2I + 1)$, they do indeed roughly scatter about a straight line through zero (Fig. 3). The theory predicts just such an intensity dependence.

3. EXPERIMENTAL PROCEDURE

The fundamental method for the spin and magnetic moment measurements is based on space quantization. Because of its spin I, the nucleus alone has a spatial degeneracy of $2I + 1$. Because of the electronic angular

momentum J, the electronic system has a degeneracy 2J + 1. In the absence of any external fields or interactions between the electrons and the nucleus, the total degeneracy is (2I + 1)(2J + 1). Because of the magnetic interaction between the nucleus and the electrons, the degeneracy between levels of the total angular momentum, $\underset{\sim}{F} = \underset{\sim}{I} + \underset{\sim}{J}$, is removed. There are either 2I + 1 or 2J + 1 of these hyperfine levels, whichever is the smaller. F takes on the values I + J, I + J - 1, $\cdots$ |I - J|, and each of these levels is 2F + 1 degenerate. On the application of a weak magnetic field, H, this degeneracy is removed, and the energy dependence of the levels is a constant plus a term proportional to H. The difference between any two energy levels is a possible transition frequency under the influence of an applied radiofrequency magnetic field, and this transition is observed, provided that it is allowed by the selection rules, and provided that the apparatus is designed to detect the particular transition involved. There are many different types of transitions, and at least three different ways of designing the atomic beam magnets to observe them. As an example, the class of transitions that involve $\Delta F = \pm 1$ is particularly suitable for precision measurements of hyperfine intervals. If no previous value of this separation is available, however, a tedious search is involved because of the high resolution of an atomic beam apparatus. A procedure that operates in stages and requires only a minimum amount of isotopes has been devised for determination of the various nuclear constants of interest. The quantities to be determined are, in order, the spin, the magnetic dipole term in the hyperfine interaction, the electric quadrupole term, the sign of each, and, if possible, the magnetic octupole term and the

hyperfine-structure anomaly. From these terms, the values of the nuclear magnetic moment and electric quadrupole moment can be determined by ratios. The sequence as described requires monotonically increasing precision. As an example, the alkalis have a $^2S_{1/2}$ ground state; therefore, the quadrupole and octupole terms vanish identically, and there is no hope of measuring these moments in this atomic state. Since the hyperfine-structure anomaly may be as high as several percent, there is no point in measuring the dipole interaction with extreme accuracy for the purpose of determining the nuclear magnetic moment by comparison with a known isotope. However, in order to determine the sign of the interaction, it is generally necessary to measure the hyperfine-structure constant to a greater accuracy to check the consistency of the assignment. As an example of some aspects of this technique, we consider Cs^{132}. The spin of this isotope is 2. Figure 4 is the plot of the energy levels versus the magnetic field. The energy is in units of a, the hyperfine-structure constant, and the abscissa is the dimensionless quantity,

$$X = \frac{(-g_J + g_I)\mu_o H}{ha(I + \frac{1}{2})} . \qquad (g_J < 0 \text{ for electrons}) \qquad (1)$$

The Hamiltonian representing the interaction between nuclear spin and electronic angular momentum, and the interaction with the external applied field, is

$$\mathcal{H} = a\underset{\sim}{I} \cdot \underset{\sim}{J} - \frac{g_j\mu_o J_z H}{h} - \frac{g_I\mu_o I_z H}{h} , \qquad (2)$$

where $\mathcal{H}$ and a are in frequency units. The Landé factor g_J is known very accurately from atomic beam experiments on the stable Cs isotopes and is

very nearly equal to -2; g_I is small, ~ 1/2000; a is approximately several kilomegacycles per second. Initially, in the weak field, or Zeeman region, the levels vary linearly with H. The corresponding frequency differences between adjacent upper levels, F = 5/2, $\Delta F = 0$, and $\Delta M = \pm 1$, are equal to one another and to approximately $\frac{2}{5}\mu_o H/h$, or about 0.5 Mc/sec/gauss. The field is too weak to decouple the nucleus from the electrons, but it does cause a precession of the entire system due to the torque on the total dipole, which is why all the upper frequencies are the same for low field. For a free electron (I = 0), this frequency is $g_J\mu_o H/h$ or 2.8 Mc/sec/gauss. The nucleus adds inertia to the system because of its angular momentum, but makes a negligible contribution to the torque; therefore the precession frequency is reduced. The exact expression for $^2S_{1/2}$ states to first order in the field for the upper levels is

$$\nu_o = \frac{g_J\mu_o H/h}{2I + 1} - \frac{2Ig_I\mu_o H/h}{2I + 1} . \qquad (3)$$

The last term is negligible for initial measurements in the determination of the spin. If a resonance can be observed at low enough values of H, the spin is determined by the factor $(2I + 1)^{-1}$. The advantage is the discreteness of the search. The resonance of the carrier or known isotope is used to calibrate the field, and the position of possible resonances for different spins can, in general, be discretely predicted and compared. Once a spin resonance is located and verified, an estimate can easily be made of the hyperfine interval, $\Delta\nu$, the difference in energy between the upper and lower F levels: $\Delta\nu = [(2I + 1)/2]a$. This is done by following

the resonance to a field that will introduce a small deviation from the linearity expressed by Eq. (3). This term is, in general, quadratic and is due to the incipient decoupling of I and J by the external field, and is

$$\nu \cong \nu_o + \frac{2I\nu_o^2}{\Delta\nu}, \tag{4}$$

to second order in the field. The shift, $2I\nu_o^2/\Delta\nu$, gives a rough estimate of $\Delta\nu$, and this shift can be increased by increasing the frequency until $\Delta\nu$ is determined to any reasonable desired accuracy. Since this procedure is capable of determining $\Delta\nu$ to 0.1% or better if necessary, one must use the exact solutions for the Hamiltonian, including the terms in g_I; g_I itself is estimated from $\Delta\nu$ by comparison with a known isotope and the relation[17]

$$\Delta\nu_1/\Delta\nu_2 = (2I_1 + 1)g_{I_1}/(2I_2 + 1)g_{I_2}. \tag{5}$$

For alkalis and other $^2S_{1/2}$ states, this formula is good to about 1%, which is more than enough for the small correction it affords. The deviation of this relation from equality is a measure of the finite distribution of the nuclear magnetism and is known as the hyperfine anomaly. However, the sign of g_I is not determined, and the usual method is to check the constancy of the calculated $\Delta\nu$ versus H for assumed $g_I > 0$ or $g_I < 0$. If the resolution of the apparatus is sufficiently good, there is enough discrimination to tell the sign.

So far, nothing has been said about the apparatus needed to observe this resonance. A particular arrangement of magnets in an atomic beam apparatus is used that results in what is inelegantly called a "flop-in"

trajectory rather than the original "flop-out" design. Reference to Fig. 4 shows the rather interesting phenomenon that the particular level corresponding to the most negative value of $M = -I - J$ (e.g., the level -5/2 in the diagram) varies exactly linearly with H and "crosses the diagram" to take its place with the levels $m_J = -1/2$ in the Paschen-Back region. The level immediately above, $M = -I - J + 1$ (e.g., -3/2 in the diagram) eventually winds up with the $m_J = +1/2$ group. The slope of these lines at a particular magnetic field is a measure of the force on an atom in that state in an inhomogeneous magnetic field. The two levels, $M = -I - J$ and $M = -I - J + 1$, therefore have equal but opposite forces exerted on them in the same inhomogeneous field, providing only that the field is sufficient to decouple the angular momenta completely as in the Paschen-Back region. Figure 5 is a schematic of an actual apparatus. The atoms leave the source and pass between the pole tips of two successive magnets that supply the field necessary to cause the decoupling and the inhomogeneity to deflect the atoms. These magnets are called the "A" and "B" magnets respectively, and are so arranged that the fields of each and the field gradients of each are in the same direction. The apparatus then acts like a Stern-Gerlach system in the first instance. The atoms are deflected either to the left or to the right, depending upon the sign of m_J, and there is no signal at the detector. Between the two deflecting magnets there is a uniform field, the "C" field, which is normally at just a few gauss for spin determinations. The resonance takes place in this field via a small perturbing rf field induced by a pair of wires, generally called a "hairpin", running parallel to and near the beam. If the

resonance condition is met, transitions are caused between the two states, $F = I + J$, $M_F = -I - J$ and $M_F = -I - J + 1$. The result is that the moments of the atoms initially in these states may be reversed in sign in the B field with respect to the A field, and these atoms will go in a sigmoid path and be refocused onto the detector yielding a signal. The result is a positive signal compared to no signal, which gives far better statistics than the flop-out method in which the indication is a reduction in beam intensity. This is an essential feature in experiments involving trace amounts of material. It is important to note that the matching and stability of the A and B fields can be quite crude without too much effect on the efficiency of the apparatus. The C field must be stable within a fraction of the line width, and since this may be as low as 20 kc/sec, this is often a stringent requirement. Since only two levels out of a total of $2(2I + 1)$ are involved, the best fractional intensity obtainable is $(2I + 1)^{-1}$, and because of various losses, about one-quarter of this is actually realized.

The final identification of the radioisotope is usually made certain by a determination of the half-life of a collected resonance sample. Figure 6 shows the results of the decay of a Cs beam produced by alpha particles on Xe. The undeflected-beam decay curve is composite, with at least three components. Three of the spin buttons collected showed counts that were above background and each decayed with a half-life characteristic of a known species. Thus the spins of Cs^{129}, Cs^{131}, and Cs^{132} were measured. Cs^{129} and Cs^{131} had been measured earlier after having been produced by different methods.

4. CONSIDERATIONS IN APPARATUS DESIGN

The design of an atomic beam apparatus for the study of radioactive species should clearly involve the optimization of the transmission which is the total number of atoms from the source that reach the detector. This is a different condition from the many that are usually employed, so a brief description is required here. The apparatus designed and constructed at Berkeley will be discussed as an example. It is more fully described in the thesis of R. A. Sunderland, and the calculations that follow are abstracted from more complete accounts in his thesis. The apparatus being considered is to be designed so that the largest possible fraction of the manufactured atoms is available at the detector. The C field will therefore be made as short as possible, thus sacrificing ultimate precision, and dead spaces between the source, the magnets, and the detector will be held to a minimum. In order to obtain a first idea, it will be assumed that the C-field length and the dead spaces are negligible. The deflection of an atom from a straight-line path is then

$$\delta y = \frac{1}{2} \frac{\mu \ell^2 \partial H/\partial y}{Mv^2}, \qquad (6)$$

where δy is the deflection perpendicular to the axis of the apparatus; μ is the effective moment of the atom; $\partial H/\partial y$ is the perpendicular magnetic field gradient; ℓ is the length of the deflecting field; M is the mass of the atom; and v is the velocity of the atom. For $\ell^2 = 1000$ cm^2, $\partial H/\partial y = 6000$ gauss/cm, $T = 1000^\circ$K, and $\mu = 10^{-20}$ erg/gauss, the deflection is 0.3 cm. In what follows, numerical factors of the order of unity will be discarded. If T is the source temperature, $Mv^2 = kT$ for an average atom.

The field inhomogeneities are developed by pole tips which are cylindrical arcs. If the radius of one of the pole tips is a, we can assume $\frac{1}{H}\frac{\partial H}{\partial y} = \frac{1}{a}$, therefore

$$\delta y = \ell^2 H\mu/kaT. \tag{7}$$

If the exit slit is of height h and width w, we may assume, dimensionally, that the source slit and width are proportional to h and w (and, in fact, h, w, and δy must be proportional to a in an analysis of this type). The total number of atoms that reach the detector is then (remembering that $\ell \propto h \propto w \propto \delta y \propto a$)

$$N \propto \frac{\rho h^2 w^2}{\ell^2} = \frac{\rho \ell^2 H^2 \mu^2}{k^2 T^2}. \tag{8}$$

Here ρ is the number of atoms per cm^3 in the vapor phase in the oven. For an experiment with an unlimited supply of material, the maximum permissible $\rho \propto 1/w$ by the Knudsen condition. This is the condition that the slit width should not exceed the mean free path in the oven, so that the condition for pure molecular effusion can be maintained. In this case, N varies as the length of the apparatus. The only limit to the increase in ℓ is the scattering of the beam atoms by the residual gas and the expense. In practice, it has seldom been necessary to consider an apparatus of length greater than 2 meters, and most of this length is due to the C field needed for high precision. In the situation in which trace amounts of material are involved, the total amount of material is fixed and all of it is needed. This is the case at hand. The proper condition is that ρhw equal a constant. Then we find

$$N \propto H\mu/T. \tag{9}$$

This means that the efficiency of the apparatus is independent of its length. The apparatus can be made quite short until the dead spaces and the finite C field introduce other limiting factors. A convenient overall length is about 75 cm. Of course, the half-life of the material also enters these considerations. As the apparatus is shortened, the rate of efflux is decreased and the carrier needed must be decreased. This can reach the point at which the amount of carrier is too small to be successfully handled.

The foregoing analysis was carried through on the assumption that the ratios of all lengths were held constant. There are other considerations, however. Provision must be made for a collimating slit and for a stop wire. The latter is an obstacle that just blocks the direct beam from the detector. It prevents the fast, relatively undeflected tail of the Maxwellian distribution from reaching the detector. Furthermore, the ratio of the A to B magnet lengths need not be unity. In fact, if the A magnet has all its dimensions reduced, it remains equivalent in focusing power and transmission, as the above analysis showed (including the necessary reduction in source dimensions). If carried to the limit, this means a factor of two in length for the total apparatus and therefore a factor of four in transmission. In practice, a factor of three can be obtained. A factor of this size is not trivial when viewed against cyclotron time and counting time. One pays for this improvement with a higher source pressure. For the alkalis, this represents a reasonable temperature rise. For low-vapor-pressure substances, such as U, Pu, Am, Np, Th, etc., the necessary rise in oven temperature is barely tolerable, and the improvement in intensity

probably is not worth it. Under these conditions, the stop wire allows only about one-half the atoms through that undergo a change of effective moment of 1 Bohr magneton. The transmission of such an apparatus is about 10^{-5} when a cosine distribution is assumed. Channeling the source never seems to produce the improvements expected from the simple theory and, further, is less effective for material of short half-life. The exit slit is 0.1 by 0.5 in., and the source is 0.002 by 0.080 in.

The one remaining question of importance is the length and value of the rf field. A typical beam velocity is 50,000 cm/sec, and if the hairpin is 1 cm in length, the transit time is 1/50,000 sec, given a resonance band with 50,000 cycles/sec. At a field of 10 gauss, a nucleus of spin I = 0 in an alkali atom resonates at 28 Mc/sec, and this band width will just resolve spins in the neighborhood of I = 50. Such resolution is far more than necessary, but usually the C field is not sufficiently uniform to attain ideal resolution, and resolutions of 200 kc/sec are more common. This is still more than adequate. The strength of the rf field is determined as follows. The system precession at resonance about the C-field direction can be viewed from a rotating coordinate system that has the same velocity and sense of rotation. In the rotating system, the angular-momentum vector appears stationary, and the applied magnetic field is taken as zero. Such is the essence of Larmor's theorem. The rf field, which appeared as a rotating vector perpendicular to the C field in the laboratory system, is now stationary if the oscillator is tuned to resonance. In this coordinate system, the spin will precess about the perturbing field H' with the Larmor frequency corresponding to H'. If the strength is just right,

it will make just one-half revolution in the time the atom spends in the rf field. Denoting this time by Δt, the condition for a precession of π is

$$\frac{2}{2I+1}\,\frac{g_J \mu_o H'}{h}\,\Delta t = \pi. \qquad (10)$$

For spin 0, $\Delta t = 1/50{,}000$ sec^{-1}, this yields a field of 20 milligauss, a field that is very easily obtained up to quite high frequencies. For a mono-energetic beam, the resonance will decrease if this field is exceeded. In practice, the Maxwellian velocity distribution will smooth out the oscillations of the resonance versus perturbing field.

5. PROBLEMS INVOLVED IN PARTICLE DETECTION OF ATOMIC BEAMS

A. General Considerations

Only experimental methods that take advantage of particle detection can now give the high sensitivities required for the investigation of short-lived radioisotopes. Two essentially complementary techniques now being used meet this requirement; one is the detection of individual beam atoms by ionization, mass-spectrometric analysis, and subsequent detection with electron multipliers; the other is radioactive collection and detection. For the first scheme, the following limiting situation is typical: at least 10^3 positive ions must be observed at the electron multiplier to obtain a statistically significant sample of the beam; mass-spectrometer transmission is about 10%; and the fraction of beam atoms that is ionized ranges from a lower limit of 10^{-4} (with the universal electron-bombardment detector first introduced by Lew and Wessel[18]) to unity for special elements, such as the alkalis and Group IIIb series, in which Langmuir-Taylor surface ionization[19] is particularly applicable. For radioactive collection and

detection this situation is typical; a beam sample is required to have a decay rate in excess of 1 cpm and is observed in detectors with an efficiency of 25%. Under these simple assumptions the cross-over point, where one method becomes comparable to the other in sensitivity, occurs for materials with half-lives between one week and 100 years, depending upon the ionization efficiency attainable. There is, however, another consideration, difficult to evaluate, but nonetheless important. Even when there is no common stable isotope having the same mass as a nuclide to be investigated by the ion method, there is inevitably some spillover of carrier, beam contamination, and apparatus background, especially oils, into the mass channel of interest. The effect is to make the ion method somewhat less sensitive than would first appear. The present cross-over therefore occurs somewhere in the neighborhood of 1- to 100-year activities. An excellent resume of the ion system is contained in an article by King and Zacharias[16] and the publication of Lew,[20] therefore this discussion is limited to research on materials with lives of 1 year or less.

The central procedures of the radioactive method are the collection of beam material for a suitable integration time and the detection of the sample decay by an appropriate counter. Let us first consider the conditions under which the material available may be most effectively utilized. The central fact is that if reliable counters are available, the relative uncertainty ϵ in a beam sampling is

$$\epsilon = (b + c)^{1/2}/ct^{1/2}, \qquad (11)$$

where c is the sample decay rate, b + c is the observed decay rate, and t

the counting time. It is assumed that the background b is well known. We dispense with the b term by noting that in most instances an efficient counting system with background of about 1 cpm or less can be found (more on this point later), and that 1 cpm is also about the lower limit of decay rate set by the time required to count down the results of a run and by the investigator's patience. Thus any discussion of radioactive detection rather naturally separates into two categories; into the first class fall all materials with half-lives of several days or more, and into the second go isotopes with shorter lifetimes. For investigations in the first class, the rate of progression of research is strictly proportional to the rate at which the beam exposures can be counted, since the exposures themselves can be taken at essentially any rate, and since the investigator will have sufficient time to analyze the data and decide on an optimum search procedure. The rate at which exposures can be counted is, from Eq. (11),

$$dN/dt = \frac{n\epsilon^2 c^2}{c + b} \approx n\epsilon^2 c, \qquad (12)$$

where n is the number of counters used, ϵ is a prescribed relative uncertainty in the beam sampling, c is the net counting rate of beam samples, and b the counter background. The total activity available for the experiment will invariably be limited by availability, expense, or health precautions, and the counting rate c will be some appropriate apparatus constant times the total activity divided by the number N of exposures required to execute the experiment. Thus perhaps a more illustrative form of Eq. (12) is

$$1/\tau = 2\eta n\epsilon^2/N^2, \qquad (13)$$

where τ is the counting time required for the entire research project, and Q is proportional to the total activity. Note that there is a large premium connected with optimum search procedures, which reduce N, and with increase of the allowable relative uncertainty in the individual resonance points. In addition, the counter effect occurs as the first power of the number of counters.

We now consider investigations falling into the second class, where one deals with materials in which the lifetime itself is a major factor in the control of exposure and counting procedures. Here the progress of the research is taken to be proportional to the number of resonance points that can be obtained per run, although other considerations, such as the fact that no extensive data analysis is possible during the course of the run, also play an important role. The maximum counting time per resonance point is $\tau n/N$, where N is now the number of resonance points taken during the run, τ is a characteristic time which is essentially the half-life, and n is the number of counters. But, as before, the individual counting rates are Q/N, so for this case, assuming $b \ll c$, we find

$$N = \epsilon(Qn\tau)^{1/2}. \qquad (14)$$

Although this result is formally like Eq. (13), the physical situation is quite different, for τ is now fixed. First of all, there is, under these circumstances, little to be gained from large numbers of counters, quite aside from the fact that in the interest of reliability and uniformity each beam exposure should be counted in each counter, so that the dead time involved in switching samples would become high (particularly for half-lives of less than a few hours). Also interesting is the fact that

N goes only as the square root of Q_T, which essentially measures the total radiation taken by the investigator. Because the run generally extends over the entire lifetime of the material and little time is left for calculations, the principal profit is in precalculation of situations that may be of interest during the course of the search procedure.

We now consider a question pertinent to either class of investigation -- indeed, to any search procedure -- namely, what method of resonance coverage, i.e. spacing of resonance points, will lead to the fastest convergence from an initial uncertainty σ in a parameter such as g_J or the hyperfine structure, whose effect on the resonance frequency is controllable (e.g. by the strength of the static magnetic field), to a final uncertainty σ'. Assuming that the assigned uncertainty in each resonance center is the interval between resonance points, and that at each stage the effect of the desired parameter on the resonance frequency is multiplied by a factor n, the total number N of points needed for the procedure will be approximately given by

$$\sigma/\sigma' = n^{N/n}. \tag{15}$$

Minimizing N with respect to n, we find that n is ideally e = 2.7 with a flat maximum between n = 2 and n = 5. The effect of this consideration, in combination with the practical ones of field drift and time spent in adjusting the field, is to make an optimum procedure one in which the effect of the parameter is increased by a factor of 3 to 6 per stage and in which resonance points are separated by intervals of 1/3 to 1/2 the line width.

Once the material has been produced, chemistry must, in general, be

performed to separate the product from the few grams of target material and to put the product in a suitable state for production of an atomic or molecular beam. The range of such "chemistry" procedures is limited only by the ingenuity and resources of the investigator. The methods are thus best illustrated by examples.

The central theme of all such processes is that between 10^{10} and 10^{16} atoms must be transferred under conditions of high radiation level to the apparatus in which the beam is to be produced under controlled conditions. The natural procedure for n - γ reactions is to bombard the pure element, which is then placed in bulk quantities into an appropriate source. Investigations using this procedure have principally been carried out by Goodman and Wexler[21] and by Smith.[22] For cyclotron bombardments or secondary reactions in piles involving a product that is not the same element as the parent, the opportunity for controlling the quantity of the carrier is a very considerable advantage, as is discussed below. Therefore, in the investigations of cesium and rubidium[23, 24], the target is not an alkali bromide but an alkaline earth bromide, and chemical separation of the target and the product is made, with a suitable quantity of carrier whose primary purpose is to prevent losses of the minute quantities of the radioisotope. The final product form is rubidium bromide or cesium iodide. The alkali halide is then reduced, in the oven, by calcium at a temperature of about 400°C. The chemistry of these two alkalis might have been done, however, in any one of the following ways. The halide nitrate could be produced in the chemistry process, the nitrate put in the oven, and then successively decomposed first to the oxide and

then to the alkali metal at high temperatures; or the azide of the alkali could be produced in the chemistry process and then decomposed directly to the metal in the oven. The gross target material, barium bromide or iodide plus carrier plus radiobromide or iodide, might be placed in the oven and the entire load reduced by calcium at an appropriate temperature.

In investigation of the neutron-deficient thallium[25] isotopes, it was found that chemistry procedures took entirely too long for the investigation of the 1- and 2-hour isotopes, therefore a different procedure was used. The thallium was evaporated (Fig. 7) from the gold target into a small cup with an appropriate amount of carrier thallium. This material was then placed in the oven and a beam made of thallium metal. Again, however, one alternatively might have put the gold target directly into the oven and evaporated out the thallium.

In gallium investigations,[26] it was discovered that the activity coefficient of gallium in copper metal is so small that one is unable to separate gallium and copper by vapor pressures, even though the ratio for the free metals at the melting point for copper is $10^3/1$, and as a result a long and involved chemistry process is required.

B. Oven Channeling

A possibility not heretofore mentioned is that the fraction of beam atoms that reach the detector, the effective transmission, may be materially increased by the use of long thin tubes or channels at the source orifice to reduce the fraction of beam atoms issuing at large angles to the beam direction. This technique has been extensively used by the MIT group in their investigations of the rare and long-lived alkalis and halides.[16]

They report typical forward effusion gains as large as several hundred from the channeling process. This technique is not of particular importance for the investigation of short-lived (less than 1 day) isotopes for the following reasons. First, in this case, one must effuse the entire activity in a period of an hour to a day. With a standard oven geometry, only a few hundred milligrams may be effused in this time. If, now, the the source is channeled by a factor K, the total effusion rate is decreased by a factor K, and the source pressure must also be reduced by about a factor K. Therefore, the total quantity of material that can be effused per unit time is decreased by a factor K^2. Absorption of the beam material by the walls of the source and inevitable sample contamination resulting from hurried chemistry set an effective lower limit of about 1 mg on the carrier; therefore, in this case, the maximum allowable channeling gain is about 20. But, from Eq. (14), the effect on the overall research picture will be a factor of only 4 or 5. An effect this small does not justify the additional line-up problems and increased probability of failure of a run. On the other hand, for moderately long-lived materials (greater than 1 day), channeling is desirable, for the chemistry procedure can be made arbitrarily clean, ovens can be properly outgassed before use, and furthermore, the effect on the research goes as the first power of the channeling gain. (See Eq. 13)

C. Beam Collection

A general problem uniquely associated with the radioactive method is the collection of the beam on some surface in such a manner as to be useful later for counting. While this process need not necessarily be 100%

efficient, reproducibility is highly desirable.

Bellamy and Smith, in their parent investigations of alkalis by the radioactive method, have used a scheme where the beam is first ionized and then attracted to a brass surface by a potential of several hundred volts.[22] It is found by Hobson and Hubbs that the retention by a brass surface for rubidium ions, but at ion currents 10^2 lower than that reported by Bellamy and Smith, is about 1% for ion energies between 10 and 1000 ev.[27, 28] The retention rises at either end of this range. It is known from other sources that the retention of aluminum for 3-kev ions in this mass region is essentially unity.[29] It is also found by Hobson and Hubbs that the retention of surfaces for thermal rubidium atoms is similarly complex. A tabulation of their data, some of which were taken only once, is given in Table II. The data were taken with a tracer technique, whereby the counting rate of samples was compared with the integrated current to a surface ionization detector, from a beam whose ratio of carrier to active rubidium is known. Because of the well-known difficulties in the precision measurement of the latter ratio, the results are normalized to unity for sulfur. (The value obtained from the measured activity ratio was about 125% for the retentivity of sulfur.)

On the other hand, Goodman and Wexler find large and reproducible retention for thermal cesium and indium atoms falling on a brass surface at liquid nitrogen temperature,[21] and Hamilton et al. find a similar situation for beams of copper and gold on surfaces of copper.[30] The Berkeley group finds that sulfur surfaces give reproducible and probably unit retention for beams of cesium, rubidium, potassium, thallium, gallium,

Table 2. Retentivity Coefficients

1.	Brass	$\alpha = .10 \pm .01$
2.	Silver	$\alpha = .07 \pm .01$
3.	Platinum	$\alpha = .24 \pm .02$
4.	Tungsten	$\alpha = .21 \pm .03$
5.	Sulphur	$\alpha = 1.0$

indium, copper, silver, gold, and neptunium. They also find flamed platinum surfaces to be equally effective for beams of thallium, plutonium, uranium, thorium, americium, protactinium, and curium. Difficulties are found by this group in the collection of bismuth, iodine, and bromine, and by Cohen in the collection of astatine.[31] Lipworth[32] et al. have tried several surfaces, including platinum and sulfur, and find a silver surface to be the only acceptable one for the collection of iodine and bromine and to give retention that is less than unity and not highly reproducible, even under conditions where the silver surface is deposited by vacuum evaporation, removed from the evaporator, and stored under mechanical pump vacuum until ready for use.

In summary, only sulfur is found to give uniformly acceptable retentivities for all the electropositive elements tried. This is presumably the result of a negative bonding energy for these materials and of the fact that sulfur forms in air a surface layer of acid which is rapidly pumped off in vacuum. The effective vapor pressure of solid sulfur at room temperature is about 10^{-9} mm Hg; therefore the lifetime of a surface atom is more than an hour. A corresponding "universal" surface for electronegative elements has not yet been found.

D. Beam Detection

The cross-sectional area of an atomic or molecular beam is typically 0.1 cm^2 or less. This immediately allows the investigator who takes advantage of this fact to design counting systems having backgrounds far below the average levels encountered in nuclear physics research. While each nuclide presents unique opportunities for the design of low-background

and high-counting-efficiency systems, the attention required by other phases of the research is such that we have standardized on three counter types (described below), which detect essentially any decay with reasonable efficiency and with background less than 5 cpm.

(1) Counter for X-rays, Low-Energy Gamma Rays, and Beta Particles. The study[33] of Rb^{81} first prompted the development of high-efficiency, low-background counters to detect K x-rays (15 kev) emitted when the material K-captures or internally converts. The product of the development is equally useful when decay goes largely through beta emission or when there is a prominent gamma (with energy below 150 kev) in the decay scheme.

The counter and appropriate collecting surface are shown in Fig. 8. The counting head is a brass cup that fits over the face of the photomultiplier, and into which are cemented the gamma window of 0.5-mil 2-S aluminum, the crystal, 0.25 by 0.5 inch by 1 mm, and the quartz light window. Care is taken to prevent a film of the cement, Myva Wax, from covering either face of the scintillator. The gamma-window transmission is approximately 90% for rubidium K x-rays; thin-film techniques might well be used to extend the useful lower limit of operation to 1 kev.

Sodium iodide crystals are obtained in large blocks and split to size in a dry box with relative humidity less than 5% at 30°C. Cleavages are made by delivering a light blow to a razor blade held against the surface in the direction of a crystal plane, and acceptable fragments with no sign of fracture or discoloration are immediately sealed between the two windows to prevent surface decomposition.

Photomultiplier tubes are selected from the large number of tubes

acquired by the Radiation Laboratory (Berkeley). Initially only 5819's were available. Perhaps one tube in a thousand would meet, at that time, the requirements of less than one dark-current pulse per minute above a pulse height equivalent to 7-kev gamma rays in NaI. The acceptance factor for the new Dumont 6292's does, however, run as high as one tube in twenty. The common denominator of tubes so selected is a very large amplification for a given dynode voltage and presumably a high photocathode conversion efficiency. The relative dark-current rate is very nearly independent of dynode voltage over the normal region of operation of the tubes. High-voltage curing of the photomultipliers leads to no improvement of the noise figure.

(ii.) Operation. Counting is performed with standard Radiation Laboratory preamplifiers and single-channel differential analyzers. The only stabilization used is that of the analyzer and a Sorenson regulator, which is used to supply 110 v ac. The counter window is customarily set to accept 90% of the x-ray line, which for 15-kev x-rays results in a 100% window, i.e. from one-half to three-halves of the peak counter. Only a 50% window is required to achieve the same results above 50 kev. Under such adjustments as this, small drifts in window width and height contribute a negligible change in counter efficiency and background, a factor of paramount importance for counting periods which sometimes are as long as three months. The counters are shut down only after a counter electronics failure.

The counter head is shielded with a 2.5-inch-thick lead cylinder. For window settings as given above, the counter background in the shield

is one count per minute or less for all energy settings between 5 kev and 100 kev. Minimum background occurs in the neighborhood of 40 kev, where it is typically 0.35 cpm. Counter background outside the shield is approximately 5 cpm. Integral settings of the counter in the shield, with a threshold corresponding to 3 to 5 kev, result in approximately 8 cpm background for a situation in which all μ mesons, low-energy gamma rays, and medium-energy electrons are detected with unit efficiency. The integral contribution of dark-current pulses to counter background is, for a typical unit, approximately $10^4 \exp(-E/0.5 \text{ kev})$ cpm between 1 and 10 kev.

The scintillation counter is ineffective for low-energy β and α emitters because of the relatively thick (7 mgm/cm^2) aluminum window.

α emitters are detected in flow proportional counters large enough that the α is stopped in the counting gas. Discrimination against pulses corresponding to less than 3 or 4 Mev lost in the gas effectively eliminates all background except that arising from large showers and from α's from contaminants in the chamber walls. This latter contribution is minimized by coating the chamber walls with a heavy layer of common aquadag which has been found to contain α emitters in much lower concentration than metals. Such chambers have backgrounds of three to ten counts per hour and a detection efficiency of about 50%.

β emitters are detected in similar large-volume flow counters, or in special units with extremely small interior wall area (circa 20 cm^2). With large chambers it is again found that a layer of aquadag substantially reduces chamber background. Large units have been found to have a detection efficiency of 50% for β's above 5 kev and a background of about 3 counts per minute. The special small-volume chambers built of stainless steel typically have a background of 1 count per minute with 50% detection

efficiency for β's above 30 Kev.

6. CONCLUSION

The experimental methods outlined here are being used to materially extend out knowledge of the ground-state properties of nuclei. Since the radioactive technique is essentially universal, the detection limitations formerly imposed on atomic and molecular beam investigations have essentially disappeared for a class of isotopes considerably larger than that which encompasses the stable and very-long-lived (> 10 yr) species. Perhaps the most significant contribution to knowledge of nuclear structure has come from the enumeration of ground-state spins and magnetic moments for a large number of isotopes of the same element; most theoretical treatments of the nuclear system are considerably simplified by the fact that the "core" remains the same across such a series. In addition to this general feature of the research, results are obtained having particular and specific interest, such as the spin 1/2 of Cs^{127} and Cs^{129}, which showed conclusively the breakdown of the shell model in this region,[24] and the spin 0 of Ga^{66} that, besides being interesting in its own right, has had some import in parity experiments.[34]

Most investigations to date have concerned only the spins and magnetic moments of nuclei. Concerted efforts are now, however, being made by at least two groups in the field to initiate similar investigations into the quadrupole and octupole moments of elements for which the electronic ground state will permit the measurement of these quantitites, and to measure a related shape-dependent function, the hfs anomaly, in those elements whose electronic state does not permit. While investigations of

this type will enormously complicate the already demanding experimental procedure, the dimensions that may be added to our knowledge of nuclear properties would seem at this time to justify almost any degree of effort in this direction.

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FIGURE LEGENDS

Fig. 1. The differential cross section for alpha particles on bromine to produce rubidium.

Fig. 2. High-yield cyclotron target for production of the radio-alkalis from halide salts. The cross ribbing is for the more effective removal of the heat.

Fig. 3. The normalized peak resonance intensity of the Rb isotopes for a number of runs. The intensities do vary approximately as $2/(2I + 1)$.

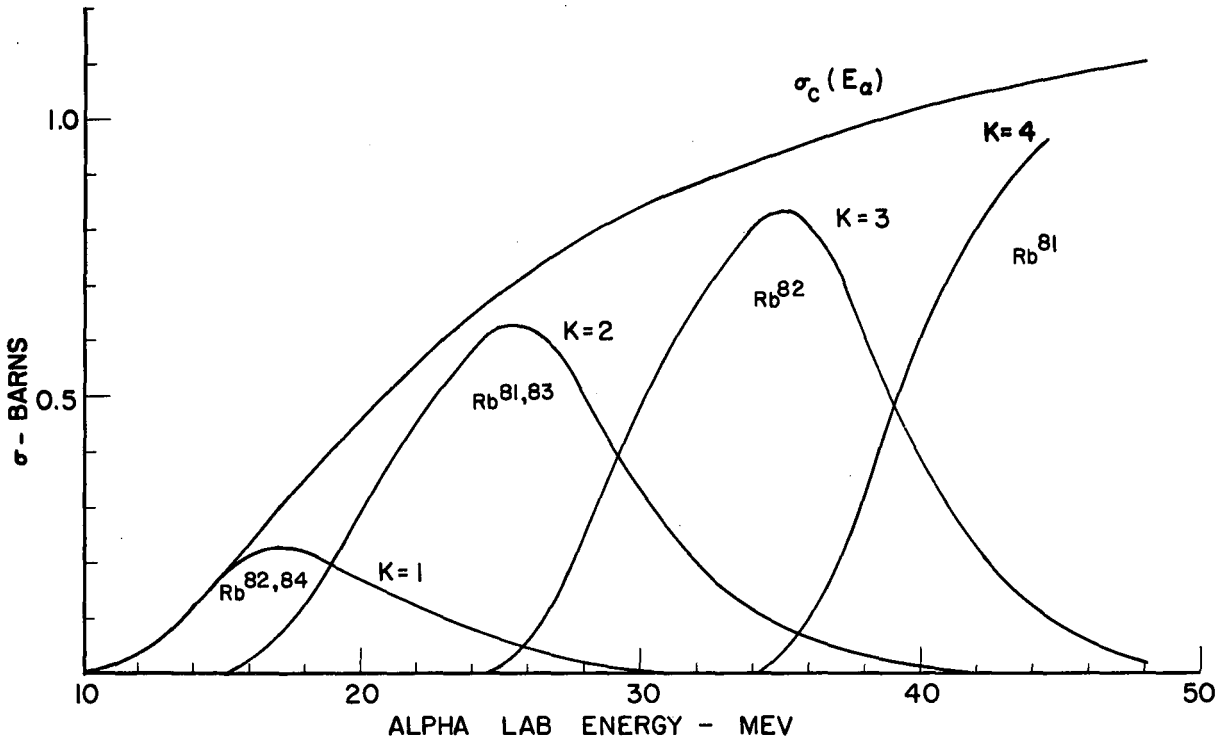
Fig. 4. The Breit-Rabi diagram for $I = 2$.

Fig. 5. A cross section of an atomic beam apparatus.

Fig. 6. The decay of three resonances corresponding to three different Cs isotopes.

Fig. 7. Detail of part of an evaporator for separating thallium from gold.

Fig. 8. Drawing of the phototube assembly for the x-ray counters. The lower part of the drawing is the sketch of the sample carriers or buttons.

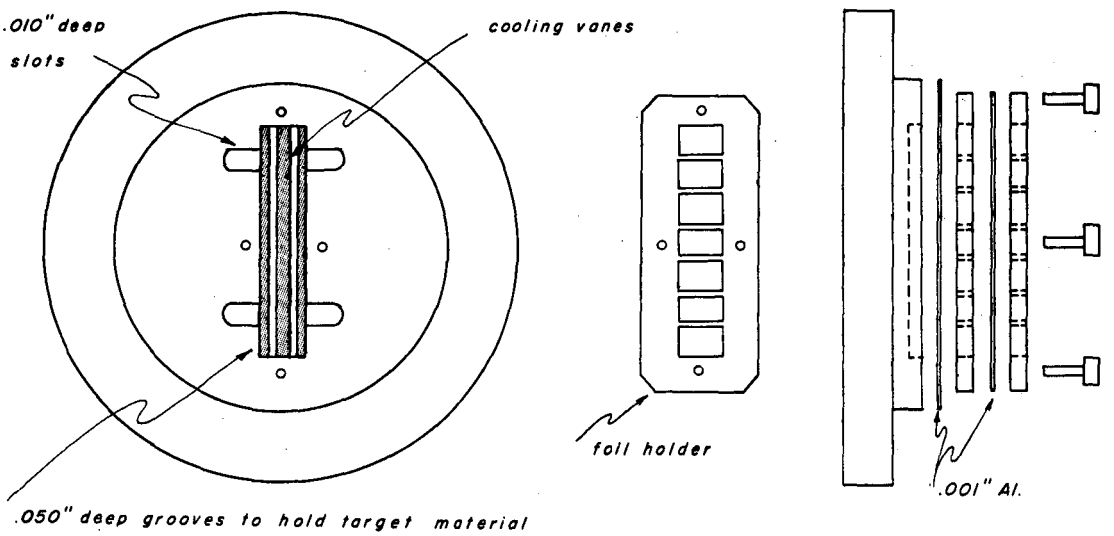


CALCULATED EXCITATION FUNCTIONS Br(α, kn) Rb

MU-11485

Fig. 1.

CYCLOTRON TARGET



MH-11482

Fig. 2.

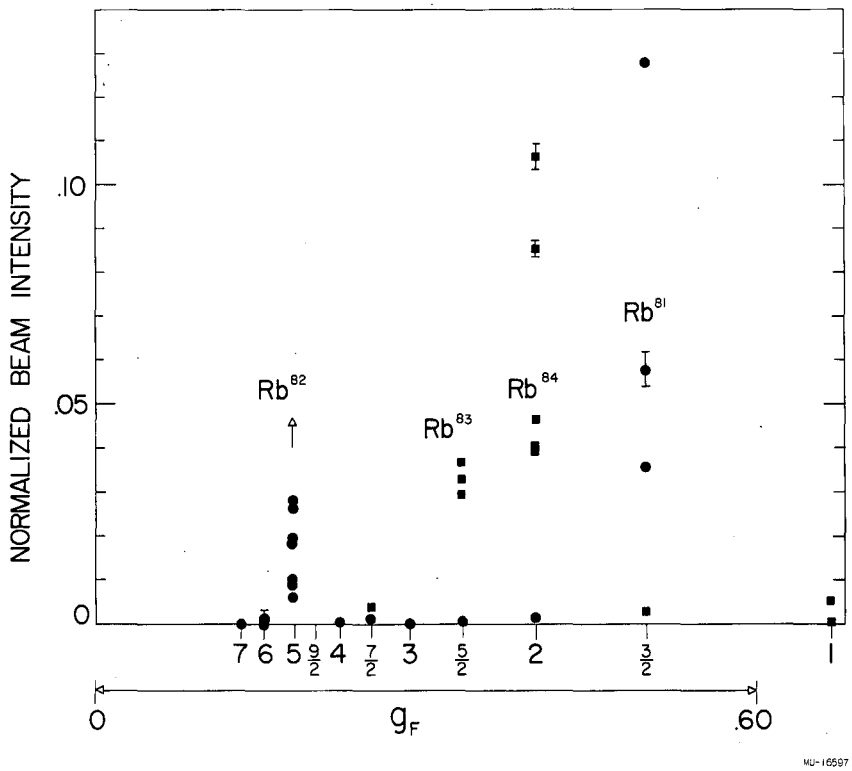
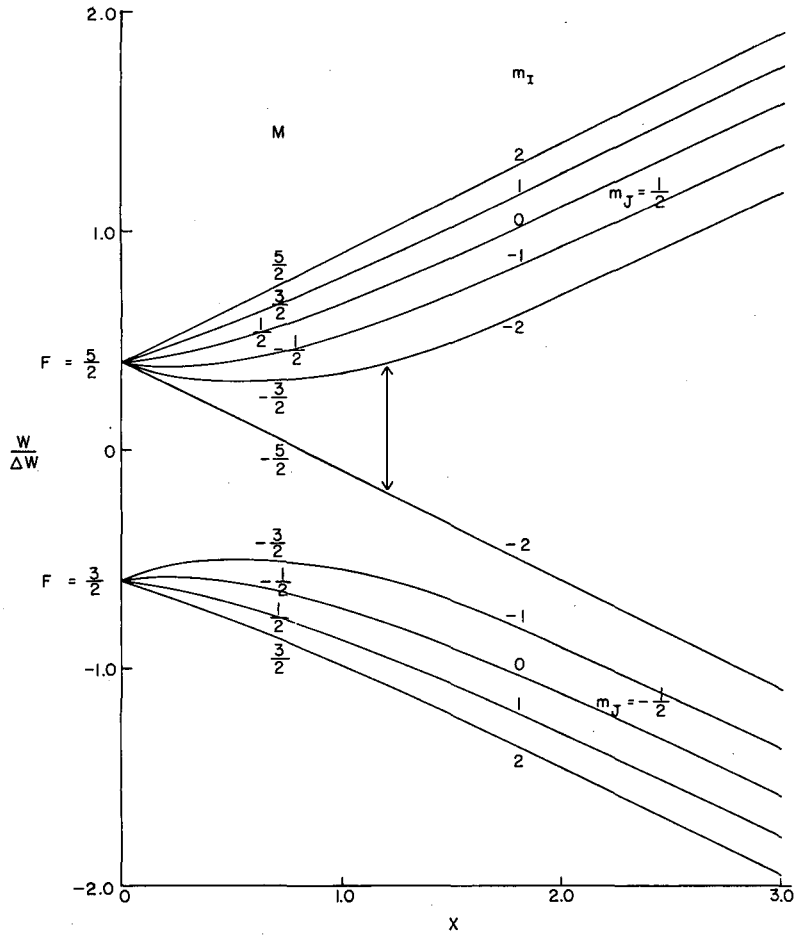

NORMALIZED BEAM INTENSITY
.10
.05
0
Rb82
Rb83
Rb84
Rb81
7 6 5 9/2 4 7/2 3 5/2 2 3/2 1
0
gF
.60
MU-16597

Fig. 3.



ENERGY LEVELS $I = 2,\ J = \frac{1}{2}$

MU-12687

Fig. 4.

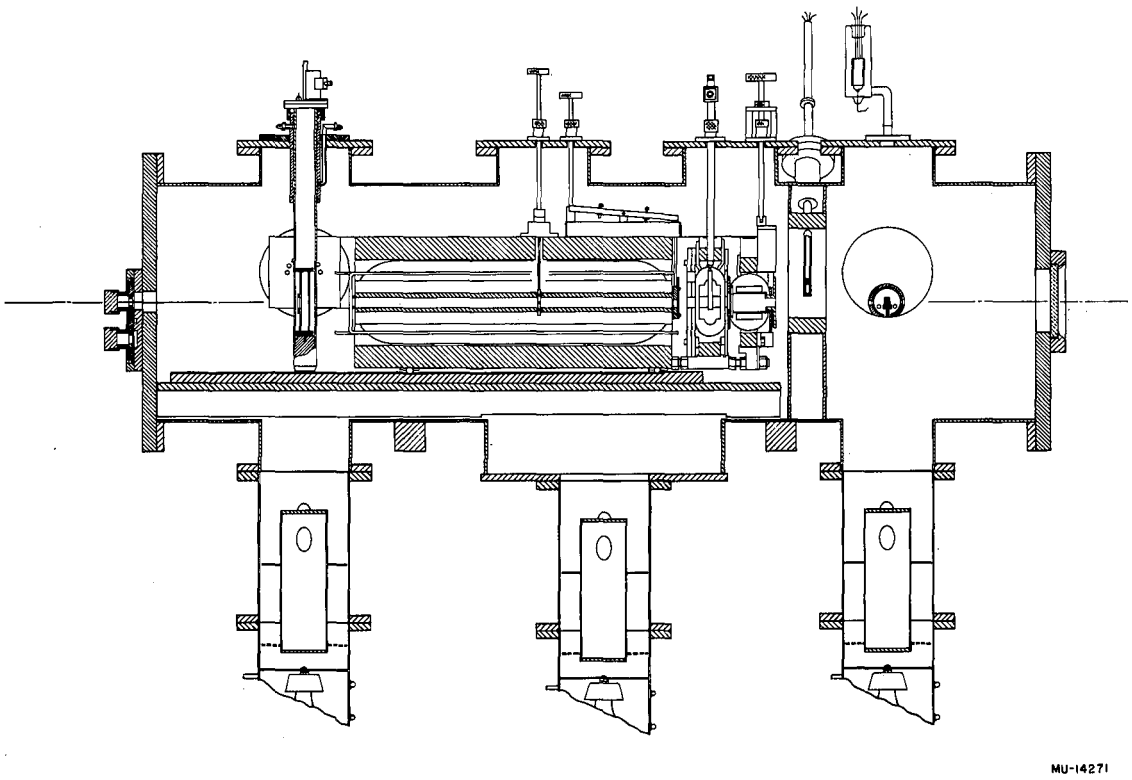


Fig. 5.

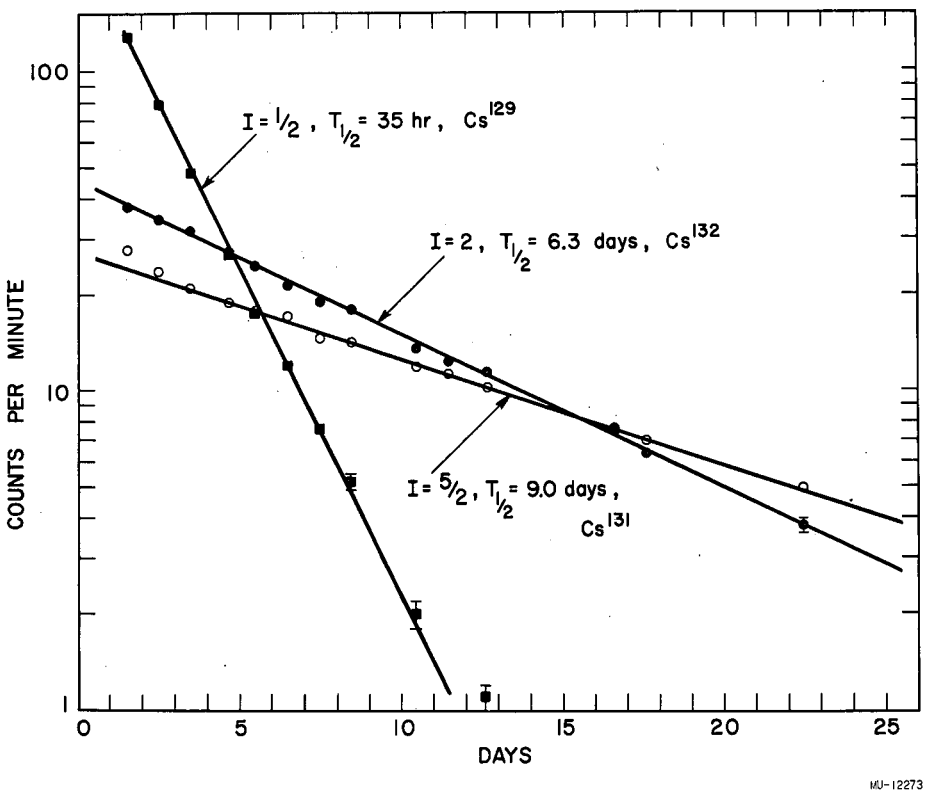
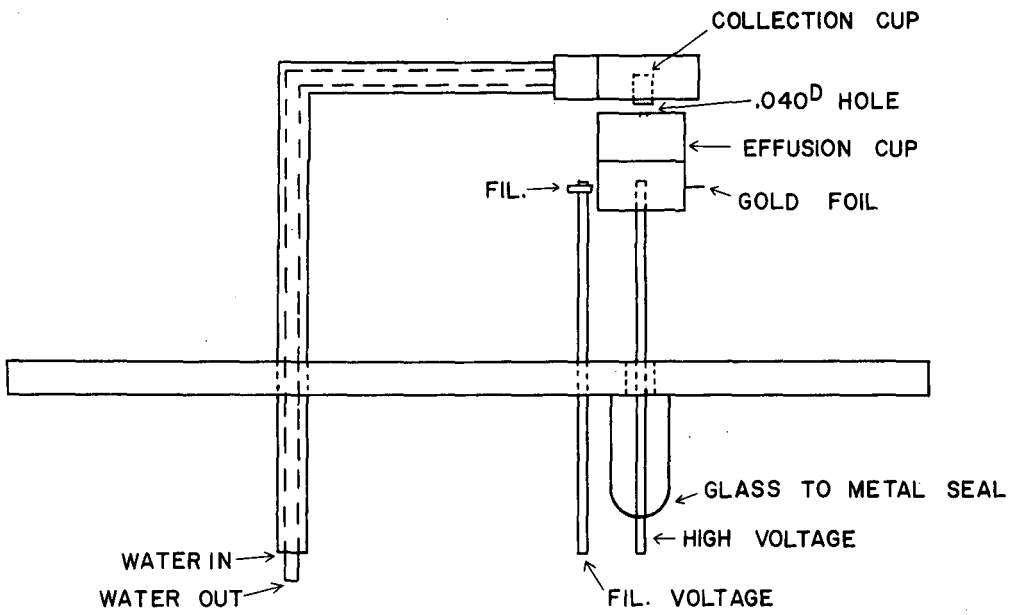

100
10
1
COUNTS PER MINUTE
0
5
10
15
20
25
DAYS
I = 1/2 , T1/2 = 35 hr , Cs129
I = 2 , T1/2 = 6.3 days , Cs132
I = 5/2 , T1/2 = 9.0 days , Cs131
MU-12273

Fig. 6



EVAPORATOR

MU-12677

Fig. 7.

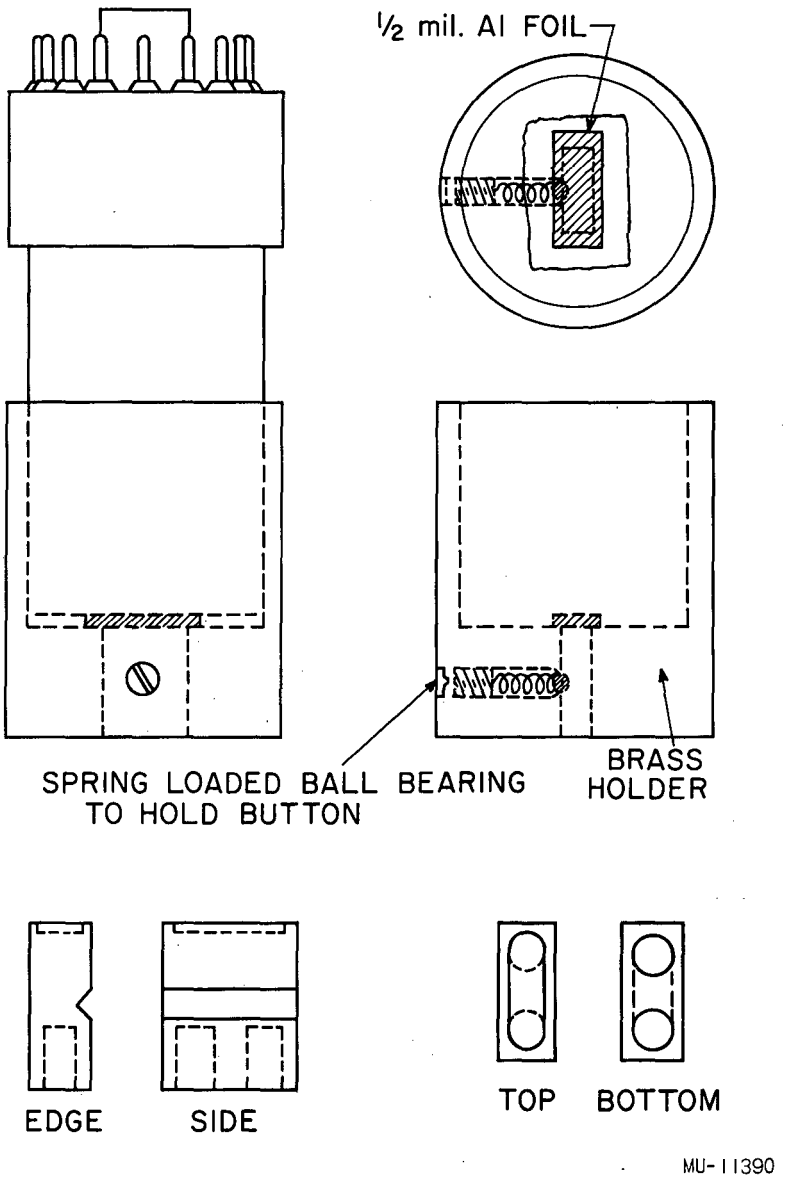

½ mil. Al FOIL
SPRING LOADED BALL BEARING
TO HOLD BUTTON
BRASS
HOLDER
EDGE
SIDE
TOP
BOTTOM
MU-11390

Fig. 8.