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ATOMIC RESEARCH ON RADIOACTIVE ATOMS (THE FIRST ERNEST O. LAWRENCE MEMORIAL LECTURE, MEETING OF THE NATIONAL ACADEMY OF SCIENCES, BERKELEY, CALIF.)

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(The First Ernest O. Lawrence Memorial Lecture, Meeting of The National Academy of Sciences, Berkeley, California)

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W. A. Nierenberg

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Atomic Beam Research on Radioactive Atoms

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FOREWORD

It is a great honor for the Atomic Beam Laboratory of the University of California to have been invited to present a review of its progress as the first Ernest O. Lawrence Memorial Lecture of the National Academy of Sciences. The larger part of this research was performed under the auspices of the Radiation Laboratory and at the specific invitation of Professor Lawrence. As most of you know, the sheer enjoyment that we in the Radiation Laboratory experience in our work derived from the spirit of infectious enthusiasm which was perhaps one of his greatest characteristics. It was irresistible - that and his keen and driving interest in basic physical research. Our group always felt this stimulus and was clearly aware of its source and was thankful for it.

I am especially grateful for the privilege of having been asked to deliver this lecture. It enables me to acknowledge publicly the great personal debt I owe to Ernest Lawrence. As Director of the Radiation Laboratory, as a senior professor, and as a neighbor on Tamalpais Road, his warm interest in my career and friendly advice were always available and greatly treasured.

INTRODUCTION

The study of the structure of the nucleus has been greatly advanced by the measurement of the angular momentum of some of the stable nuclei. This magnitude is quantized and, when expressed in units of Planck's constant divided by 2π, the maximum value of its projection along an arbitrary axis, I, is an integer or half-integer and is called the nuclear spin. The quantum mechanics of space quantization allows precisely $2I + 1$ states of orientation of this angular momentum, and the schematic situation is illustrated in the first slide. (Fig. 1.) This is called the degeneracy of the spin states. It is one of the constants of motion of the ensemble of particles that constitute the nucleus and as such is a fundamental nuclear quantity and its value can be used as a test against any special nuclear model or theory. In fact, one of the great successes of the shell theory proposed by Maria Mayer, and Haxel, Jensen, and Suess, was just this ability to predict ground state spins of nuclei. The natural nuclei scatter along the so-called stable line of the Segrè isotope chart, but most of the nuclides known are not stable -- some being deficient by as much as ten neutrons from this region of stability. It is very desirable that the spins of as many isotopes as possible be measured to provide a basis for further advances in the theory of nuclear structure. The experimental difficulties in making these measurements are formidable and arise from several sources. One is the short half-life of many of these isotopes. A typical lifetime is of the order of one hour and techniques needed to prepare a suitable cyclotron target, extract the isotope, put it in an appropriate form for the measurement and then do the measurement in a time of the order of one

hour are fairly precise and fairly delicate. Another difficult feature of the measurements involves the small number of available atoms. For these short half-lived isotopes, numbers of the order of 10^{10} to 10^{13} are available. In the past, optical spectroscopy, microwave absorption, paramagnetic resonance in crystals, atomic beams and nuclear magnetic resonance have been the principle methods of spin determination for elements which are available in quantities of the order of 10^{18} or more atoms. This is unfortunately about six orders of magnitude too large for the broader class of radioactive nuclei. This talk is primarily concerned with the solution of this experimental problem by a combination of particle detection techniques and atomic beams. The results have been successful in that in the last three years the spins of fifty-two unstable isotopes have been measured in our laboratories with half-lives ranging from 18 minutes to 25,000 years, covering a wide variety of elements from potassium to curium. In addition, the techniques often allow the determination of the magnetic moments and the electric quadrupole moments of these nuclei, and this has been done in many of these cases. These quantities are also very important nuclear constants. They represent important examples of moments, current density and charge density of the nucleus, and are shape-dependent factors very suitable for testing nuclear models. This work is continuing and is being expanded to include hyperfine anomaly studies on some of these isotopes.

The atomic beam technique is equally successful for precision measurements of similar quantities for the electronic structure of the atom. As a result, the angular momenta (in the same units of $h/2\pi$ as the nuclear spin) of the low-lying states of several transuranic elements have been determined,

as well as the electronic magnetic moment (Landé g-factor) of these states, and the results afford very keen insight into the correct ground electronic configuration and coupling schemes. For example, the results seem to have confirmed the actinide hypothesis.

THEORY OF THE METHOD AND EXPERIMENT

The atomic beam part of the experimental method for spin measurement is quite old and was developed originally by Zacharias for the measurement of the spin of K^{40}, an important problem in beta decay theory of its time, as a modification of the atomic beam resonance of Millman and Kusch, which in turn was developed from the molecular beam resonance first developed by Rabi. An atom of non-vanishing electronic angular momentum has a magnetic moment due to the electron motion and electron spin, and the angular momentum precesses due to the torque of an applied magnetic field in a manner similar to a gyroscope's motion subject to the torque of the earth's gravitational field. The frequency of this precession is proportional to the magnetic field and the coefficient is called gamma, the gyromagnetic ratio. For many of the elements, this ratio is very well known from measurements on the stable isotopes, and if we take the simple case of the alkalis, for example, which have only one valence electron with no orbital-angular momentum but only a spin-angular momentum, this precession frequency is 2.8 megacycles per second per gauss. If a weak oscillating field of the order of 100 milligauss is added to the fixed field, a resonance can take place and the precessing alkali atom will open its cone of precession when the frequency of the oscillating field matches the precessional frequency of the atom. In quantum mechanical language, the atom whose $J = 1/2$ will change from one

of its two projection states to the other. The apparatus is designed to detect this change in orientation of the alkali atom. This is the situation neglecting the nucleus. However, the electronic motion generates a magnetic field at the position of the nucleus which is of the order of several hundred thousand gauss and this interacts with the nuclear magnetic moment to form a tight coupling between the two angular momenta and yields a total angular momentum for the atom for which the symbol F is used, which is quantized as well. The nuclear angular momentum is of the same order of magnitude as its electronic counterpart and the change in the total for the system is appreciable. The nuclear magnetic moment is very small, however, and is about one two-thousandth of that of the electrons and therefore contributes very little additional to the total magnetic moment. We have, in consequence, a system of appreciably increased angular momentum, but upon which is acting essentially unchanged torque, and therefore the precessional frequency is reduced. The effect of the nuclear spin can thus be observed on an atomic scale. For example, the precessional frequency for an alkali is $2.8/(2I + 1)$ megacycles per second per gauss. The next slide (Fig. 2) is a simple table to illustrate this point. The fixed magnetic field is set to a value that will cause a resonance of 10 megacycles per second for a nuclear spin of 5/2. This would be some known calibrating isotope, say Rb^{85}. The table gives the corresponding resonance frequencies for different nuclear spin values up to values of the spin I = 13/2. The resolution of a crude atomic beam apparatus is about 0.1 mc/sec, but it can be made as low as .001 mc/sec to allow more adequate resolution for distinguishing spins, if necessary. The experimental procedure is then very simple. A magnetic field is calibrated with a known

stable isotope, usually one of the heavier alkalis which are the simplest to deal with. Then the resonance oscillator is set to a series of discrete frequencies determined by the simple indicated table for whatever spins are considered possible, and an effect will appear for one of these values if there is a suitable detection scheme.

The detection method employed is based on the space quantization diagram of Breit and Rabi. The next slide (Fig. 3) is a dimensionless plot of energy levels of an atom with $J = 1/2$ and $I = 3/2$ versus applied magnetic field. For very small values of the field, approximately several gauss, the levels vary linearly with field and the Planck condition for the transition frequency between the levels labeled -2 and -1 in the upper set is just the one we have previously described. The two groups correspond to a total angular momentum $F = 3/2 + 1/2 = 2$ and $F = 3/2 - 1/2 = 1$ and are split into five levels and three levels respectively. This is the Zeeman region. In very strong fields, of the order of several thousand gauss, which correspond to the region to the extreme right of the diagram, the electron is completely decoupled from the nucleus and its precessional frequency is that of a free electron. It has two orientations corresponding to the two parallel groups of energy levels in the diagram. Each group is composed of four levels corresponding to each of the four states of orientation of the decoupled nuclear spin. The upper group corresponds to energy increasing with field, because the electron is in a parallel state with respect to the field, and the lower group corresponds to decreasing energy and an antiparallel state. Since, in a weak field, the groups are five and three in number and in strong fields the groups are four and four, one level, $M = -2$, must "cross

the diagram". This is a unique consequence of the space quantization. Returning to the strong field situation, some consideration will indicate that an atom in one of the upper states will be urged one way in an inhomogeneous magnetic field, and an atom in one of the lower states will be urged in the opposite direction. This is the region used by the focussing parts of the apparatus. The next slide (Fig. 4) shows a schematic sketch of an atomic beam apparatus. On the left is a source of the atoms, which emerge with thermal velocities. The source may be an oven with slits to supply the vapor or a discharge tube also with slits for dissociation of gaseous molecules. The chemistry necessary to prepare the source from a cyclotron target is often very difficult and varies very widely from element to element. For the alkalis, a final reduction of an alkali halide by calcium directly in the oven can give a useful beam, whereas for some of the transuranics a conversion of the oxide to the carbide and then thermal dissociation seemed to be the only successful one tried. For separation methods in the restricted time allowed, methods such as precipitation, electroplating, high-temperature distillation, and elution columns have been used. Fairly elaborate precautions have to be taken to keep up the specific activity of the sample. A fairly complex vacuum-loading and oven-heating system is essential for rapid operations. After emerging from the oven, the atoms pass through two successive strong fields, which are inhomogeneous and in the same sense, and the strength of the fields is such that the atoms are deflected to the left or right, depending upon whether the decoupled electron is parallel or antiparallel to the field, and never reach the detector at the end of the apparatus. Between these two fields is a very

weak uniform field and the still weaker oscillatory field. If the frequency is correct, an atom that was in state -1 is now in state -2 and vice versa. The net result is that these atoms have their moments reversed in sign and are refocussed to the detector. There is a stop wire that blanks out the fast tail of atoms of the Maxwell distribution that would tend to go straight through in any event. We have achieved the desirable situation where there is no signal unless there is a resonance. This is an essential feature of the statistics where small numbers of atoms are to be detected. The usual detector is a hot tungsten surface ionizer that works very well for the alkali calibrator, but is, of course, unsuitable for most other elements and particularly the small number of radioactive atoms in the beam. The detector that is used is a collecting surface that interrupts the beam and is withdrawn through a vacuum lock and then counted by a suitable particle detector. The collecting surfaces turned out to be no trivial problem, and, after much research, it was found that sulphur was unusual in that it had close to 100% retentivity for incident electropositive atoms. This is not true for most other surfaces, such as brass, iron, nickel, etc. Sulphur is poor for halogens and there fresh silver plate was found adequate. Finally, conventional radioactive counters were found to be inadequate, since the background was too high for any reasonable efficiency. Fortunately, very special features of an atomic beam could be used. The cross section of the beam at exit is approximately a tenth of an inch by one-half inch. Furthermore, many of the interesting isotopes decay of K-capture and emit characteristic x-rays. These x-rays are absorbed in 1/2 mm of NaI or less, and so a NaI crystal counter and phototube, using a very small

crystal plate 3/4" x 1/4" x 1/16", will count just one-half the disintegrations with very low background because of its small volume. The discrimination that can be used because of the known x-ray energy keeps the background rate down and also serves as further identification for protection against contamination. The best results have been with the isotopes of Cs, where the background is one-third of a count per minute, with an efficiency of 45%. The principle factor is the 2π loss of solid angle. Small volume electron flow counters have been developed that are nearly as good for the neutron excess isotopes and the use of the alpha counters for low background work for some of the transuranics is routine.

To recapitulate, alkali atoms issue from the oven into the strong inhomogeneous A field, half are deflected to the right and half to the left, depending upon the decoupled electron spin orientation. They enter the very weak C-field region where they encounter a still weaker oscillatory magnetic field. If the frequency of this field matches the Planck transition frequency for the inelegantly-termed "flop-in" transition, the atoms of the corresponding states interchange in such a fashion that when they emerge into the B field, which is also strongly inhomogeneous, they are deflected in a direction opposite to the original deflection and are focussed to the detector.

The following series of slides will convey some idea of the experimental complexities. The next slide (Fig. 5) is a photograph of one of the four apparatuses at Berkeley designed for this research. One apparatus is primarily for condensable beams, such as Cu, Au, etc. Another apparatus is for gas sources, such as the halogens, which require a special discharge

source for dissociating the molecules. A third apparatus is for the transuranic and heavy elements, which require special source techniques of their own, and the fourth apparatus is designed for the purpose of making precision measurements to a part in 10^7 or better, so that hyperfine structure anomalies of radioactive isotopes may be studied. This particular slide is a picture of the precision apparatus. On the left is the source and special source loader. The magnets are in the vacuum envelope, but the leads are visible. On the right can be found the hot wire detector and the vacuum lock for inserting and removing the beam collectors for counting.

The next slide (Fig. 6) is an exploded view of the cyclotron target that was developed for getting maximum production of radioactive atoms where halide salts were used. Beam currents of 60 microamperes for 48 Mev alphas were achieved without destroying the target by using a double aluminum grid to remove the heat efficiently. Other targets were simple metal foils and in two other cases were gas targets. The next slide (Fig. 7) is a sketch of the x-ray counters and one of the sulphur-coated brass buttons that was found to give full efficiency for collecting atoms. It can be seen that the NaI crystal is very small by comparing it to the photomultiplier tube. The assembly is indicated on the diagram and the brass collector is fixed in place by a spring-loaded ball bearing that locks a V-groove in the side. This gives the needed reproducibility.

EXAMPLES OF RESULTS

Since something like fifty-five isotopes have been analyzed to some degree, it is impossible in this talk to cover the results adequately. The first of the experimental results will be chosen for the purpose of

illustrating the applications of the method and demonstrating some of the interesting features that arrive in the laboratory. The next slide (Fig. 8) displays the results of the simplest possible experiment that can be attempted. The substance is Ag^{110m}. It is made by putting a sample of natural silver in a nuclear reactor for several weeks. It has a long half-life (250 days) and so the experiment was leisurely performed. It is already in metallic form with carrier silver, and hence no chemistry is required. A simple tantalum oven was employed and the formation of a silver beam is well understood. It was on silver atoms that the classical Stern-Gerlach experiment was performed. Plotted horizontally are frequencies which are indicated by the appropriate resonant spin values. The ordinates are the counting rates for each of the exposures taken. The signal is high for $I = 6$ and low for the other spins. In this simple case, the signal-to-noise ratio is very good, despite the fact that the intensity of the signal is particularly low for high spins, varying as the reciprocal of $2I+1$. This can be understood from the Breit-Rabi diagram in that only two of the levels contribute to the resonance and the total number of levels varies as $2(2I+1)$, the products of the degeneracies of the electron spin and the nuclear spin states. The next slide (Fig. 9) is a plot of the full resonance line of Ag^{110m}. The experiments are usually more complicated than this; because the lifetimes are short, a chemical extraction has to be performed, and more than one isotope is produced. The next slide (Fig. 10) is the production curve for Rb^{81}, Rb^{82}, Rb^{83}, and Rb^{84} by alpha particles on bromine. It is the differential cross section with respect to energy versus the energy of the alpha particles. There are two natural isotopes of bromine in equal abun-

dance, and so all four isotopes are made; 81 and 82 in two ways. The half-lives are 4.7 hr, 6.3 hr, 83 days, and 33 days respectively. The bombarding salt is $BaBr_2$ and a separation is performed after bombardment using RbBr as carrier in controlled amounts. It is unsuitable to use RbBr in the bombardment to avoid the chemistry, because the natural rubidium would be in excessive amounts for the half-lives involved. The additional rubidium carrier increases the time for emptying the oven well past the half-life of the short-lived rubidiums. Spin buttons were exposed from I = 0 to I = 6, including the half-integer spins. Two resonances are observed when the buttons are counted immediately after exposure, one corresponding to I = 3/2 and one to I = 2. Clearly the 3/2 value is associated with Rb^{81} and the 2 value with Rb^{82}. The counting rates of the two resonance signals are plotted versus time and are shown in the next slide. (Fig. 11.) The 3/2 resonance decays with a half-life of 4.7 hr and the 2 resonance with a half-life of 6.3 hr. These were just the reported half-lives of Rb^{81} and Rb^{82} and constitute a very firm identification. As a reference, the decay of the main beam, received at the detector with the magnets off is plotted on the same curve. The half-life is 5.4 hr, representing the starting mixture of both isotopes, and the difference is quite evident. The two resonances represent a fairly complete separation of the two isotopes, and this separation is a completely independent verification of the spin assignments. This is a very valuable technique and is useful in those cases where many isotopes are present. After waiting a few days to allow the Rb^{81} and Rb^{82} to decay away completely, the spin samples were recounted and two more resonances were found, one at spin 5/2 and one at spin 5, which were assigned

to Rb^{83} and Rb^{84} respectively. This assignment was also checked out by decay. An anticipatory remark is that the spins of these isotopes and many of the others are further verified in the measurement of the hyperfine interval, which is the energy separation (which will be expressed in frequency units) between the two states, parallel and antiparallel, in zero field. The feature of spin assignment by the isotope separation has been used many times in this laboratory. An interesting case is that of Rb^{81m}, which is a 30-minute isotope. The resonance is not as clear as it might be because of the hurried technique necessary for this short half-life. However, the change in decay rate of the resonance at spin 9/2 versus the main beam is marked. It is interesting that this method is the only one for separating nuclear isomers before decay. The shortest half-life isotopes treated this way have been Rb^{81m} and Cs^{130} with half-lives of 30 minutes. This is close to the limit of present techniques, unless there are very accidental features, as was the case of Br^{80} whose half-life is 18 minutes. The next slide (Fig. 12) is the decay of three cesium resonances observed as a result of proton on xenon bombardment. Three distinct resonances were observed, which decayed with distinctly different half-lives and differently from the main beam. This is an especially fine result of the isotope separation technique. This was a particularly enjoyable result, because the Cs^{129} had been previously measured in Berkeley after having been produced by alphas on iodine; the Cs^{131} had been previously measured by Smith and Bellamy in Cambridge; the Cs^{132} was a new result.

The examples chosen have been from the simplest situations, where $J = 1/2$. This group includes the alkalis, gold, silver, copper, the lowest

atomic states of indium and gallium, and others. A more complicated situation, but potentially more interesting, occurs when the atomic angular momentum is greater than 1/2, say for example, the first excited state of Ga, which is $J = 3/2$, which is appreciably populated at the oven temperatures necessary to make the beam or the atomic ground states of the halogens where J also equals three-halves. The following slide (Fig. 13) is the Breit-Rabi diagram for this case, where I has been chosen 5 for illustration. Notice that there are four levels at zero field and four groups of levels at strong fields corresponding to the four orientations of the electronic angular momentum. The diagram is very complicated but, in the order of the levels shown, the two arrows indicate the two "flop-in" transitions that are observable in this case. Thus, while the intensity is further reduced, the observation of two resonances is even stronger confirmation of the spin. Only one of many possible examples will be illustrated. That is the measurement on Ga^{66} and Ga^{67}. These isotopes are produced by alpha bombardment of copper and the separation was performed by electroplating. The results are summarized in the next slide (Fig. 14). They are complicated by the fact that there are two isotopes and two resonances each for the $J = 3/2$ state and one resonance each for the $J = 1/2$ state, which is also present in the beam. The $I = 0$ resonance for $J = 3/2$ is very evident and must be assigned to the Ga^{66} because of the integral-even mass number assignment and the decay rate of the resonance. There is a triple numerical coincidence in that the $J = 1/2$ resonance for spin 0 coincides with both resonances for $J = 3/2$ and spin 3/2, which is the Ga^{67} spin. The $I = 0$ component was separated by decay-rate analysis, as indicated; the two

resonances for I = 3/2 separated at higher fields when hyperfine-separation measurements were made. This is probably too complicated a situation to discuss in detail in this talk, but it serves as an indication of the painstaking analysis that is sometimes needed. This particular result was interesting because Ga^{66} was the first nucleus composed of an odd number of protons and an odd number of neutrons to have its spin definitely established as zero.

The next slide (Fig. 15) is one of two compiling a brief summary of the spins and moments measured in Berkeley. There are included some spins of the stable isotopes and radioactive isotopes of the same elements which were measured elsewhere. The asterisks indicate the spins measured at Berkeley for the first time, and the dash indicates a verification of a previously measured isotope. There is not sufficient time, nor is it the purpose of this lecture, to discuss the nuclear implication of all the results, but some general results are worth noting. There is a gold isotope, Au^{194}, that is especially interesting because of its particularly small magnetic moment, namely .008 nuclear magnetons, compared to the typical value of 1. This is an interesting accidental value, because a zero-spin measurement by most methods, including the present one, really is the determination of an upper limit to a magnetic moment. In the case of Ga^{66}, for instance, this limit is 10^{-5} nuclear magnetons, but there is always an ambiguity. As another example, there are several elements that have 8 to 10 isotopes whose spins have been measured. Some of these isotopes are far removed from the general line of stability. These offer a fruitful field for comparison with theory and future possibilities for hyperfine structure

anomaly studies. The Ga^{66} spin of zero was previously noted. The next slide continues the list. (Fig. 16) The Cs^{127} and Cs^{129} results, with spins of one-half each, were entirely unexpected and have proven very difficult to reconcile with current theories of nuclear structure. There are additional groups of isotopes that have of the order of ten members included, such as silver, gold, thallium, and iodine. Astatine-211 yielded its spin by these methods, even though considerable difficulty was encountered in developing a suitable beam carrier, no natural one being available. The field of research at the end of the table is active for reasons other than nuclear as well.

MAGNETIC MOMENTS, QUADRUPOLE MOMENTS AND SIGNS

Reference has been made several times to nuclear magnetic moments determined in these laboratories. The nucleus does interact directly with the external magnetic field. This is a very feeble interaction, but its effect is often directly observed because of the high precisions obtainable. However, this method is neither suitable nor useful where the amount of material is limited. Instead, use is generally made of the fact that the hyperfine separation of the two levels in the $J = 1/2$ situation, for example, is proportional to the nuclear magnetic moment, since it involves the interaction of that moment with the magnetic field of the valence electrons. This magnetic field is essentially the same for all isotopes of the same element, and hence if the magnetic moment is measured directly for a stable isotope, say by the Bloch-Purcell nuclear induction technique or the Rabi molecular beam technique, to mention just two methods, the coefficient of proportion-

ality can be applied to the radioactive isotope to yield its moment, if its hyperfine separation can be measured. This is the essence of the Fermi-Segrè formula. The method adopted for measuring the separation is based on the fact that the frequency of transition cannot remain linear with the field as it increases, but must have quadratic and higher terms as the increasing field decouples the nuclear spin from the electronic angular momentum by competing with the internal field that couples them. In the Breit-Rabi diagram, this effect causes the increasing curvature of the energy levels with increasing field. Simply stated, we can write approximately that

$$f = f_I + \frac{2If_I^2}{\Delta\nu} .$$

f_I is the transition frequency if the coupling were very tight and the frequency were linear with the field, f is the true frequency, and the quadratic term represents the shift. $\Delta\nu$ is the hyperfine separation and is clearly correctly placed because, if it were infinite, the shift would vanish. Before a measurement is attempted, a very crude estimate of $\Delta\nu$ can usually be made. The field is set at a value where the shift is small, so that only a few resonance points need be taken. One or several will be high, giving a much better value of $\Delta\nu$ to try at a higher field, and so on. It is often the case that, with less than 50 experimental points, the hyperfine structure can be measured to about one percent and ergo the moment. The next slide is an example drawn from the research on Tl^{204}. (Fig. 17.) It is a series of four searches at increasing fields completed in one afternoon. Each graph is a plot of the counting rate of the exposed button versus the

applied frequency as the abscissa. The top line of each graph is an equivalent abscissal scale, but translated to the value of $\Delta\nu$ if the resonance were to center at that point. The rapid expansion of the scale of accuracy is evident as the field is increased. The hyperfine structure constant was determined here to better than 1%. Eventually a direct transition at weak fields was observed, and the constant was determined even more precisely. For purely nuclear theory, this additional precision is usually not warranted.

Now, when $J = 1/2$, there is only one hyperfine separation at zero field, and only one constant can be determined and that one is related to the nuclear magnetic dipole moment. If $J = 1$, there are two such differences, and therefore two such constants must be involved; the second is the nuclear electric quadrupole moment, a direct shape factor. If $J = 3/2$, there are three constants, the third is the magnetic octupole moment, and so on. This is a simple proof that a nucleus cannot display a static electric or magnetic moment greater than 2I in order of complexity. Actually, very few octupole moments have been measured in toto, the effect is too small, and they have been neglected, so far, in radioactive research. The general principle of measurement of the first two constants is the same, however, as for the $J = 1/2$ situations, although more resonance lines must be observed for accurate determination, and it has been found convenient to employ electronic computers to analyze the results. This allows greater freedom in choice of transitions. Many isotopes have been so treated, but only one example of special interest will be discussed. It developed that Br^{76}, $I = 1$, had a large enough quadrupole coefficient compared to its magnetic coefficient

that, as a result, a peculiar inversion of its levels took place, the first of this kind ever observed, I believe. The next slide (Fig. 18) shows the Breit-Rabi diagram for this isotope. The F = 1/2 level, which is normally the lowest, is now between the 3/2 and 5/2 levels, and as a result, a third transition, as indicated, is possible and was observed, thus confirming the assignments.

The only remaining important question is that of the absolute sign of the interaction, or equivalently, the signs of the nuclear magnetic moment and the electric quadrupole moment. This can also be measured by a fairly complicated expansion of the technique and requires a detailed discussion for which there is no time.

HEAVY ELEMENT RESEARCH

The heavy elements, defined as those beyond radon, have recently proven themselves to be a most fruitful field for atomic beam research. At the start of this program, little was known about the ground state properties of these atoms beyond a certain similarity to the rare earths, established by chemical methods. These methods, including magnetic susceptibility determinations, had clearly established the presence of 5f electrons and possibly the existence of 6d electrons in the crystalline state, but almost nothing was known of the configurations of the free atom and even less of the electronic coupling constants. It was generally assumed that coupling schemes and configurations would be quite similar to those in the rare earth region. The usual approach is the study of the visible and ultraviolet spectra of these elements, but these are extremely complex and have so far resisted anything resembling complete term analysis. As a result, many important

questions have remained unresolved, including the spectroscopic classification of the ground states. The method of atomic beams is very powerful in this regard in that it often results in unambiguous assignments of the electronic angular momenta and magnetic moments of the ground state and other low-lying states. The magnetic moments can be measured to close to four significant figures and the accuracy permits estimates of the deviation of the system from certain ideal assignments which represent conventional limiting situations. In technical terms, the accuracy permits detailed estimates of the nature of the coupling among the electrons and the effects of configuration mixing. It is convenient to discuss the electronic magnetic moment in terms of the dimensionless Landé g-factor, which is essentially equal to 2 for the spin of a single electron with no orbital angular momentum. In this notation, the electronic gyromagnetic ratio of an alkali is -2, that of the $J = 3/2$ state of a halogen is $-4/3$, of the $J = 1/2$ state $-2/3$, and so on.

Since the start of this research, the atomic beam resonance method has been used to measure the ground state angular momenta and Landé g-factors of one or more low-lying electronic states in ^{91}protactinium, ^{93}neptunium, ^{94}plutonium, ^{95}americium, and ^{96}curium, with the result that more is now known about the ground states of the free heavy element atoms than about those of the free rare earth atoms. Before discussing the general nature of the results obtained, we will digress to discuss some of the experimental problems that had to be overcome. First and foremost, is the problem of beam production. The oven source temperatures required vary from 1900° Kelvin for curium to 3100° Kelvin for protactinium. The curium and protactinium

beams are formed by mixing the nitrate with carbon in tantalum ovens and raising the temperature gradually. The nitrate becomes the oxide and then the carbide,and finally the carbide decomposes at a rate to give a sufficiently reliable beam. The plutonium and americium are evaporated directly as the metal, the former in a tungsten oven, the latter in a tantalum oven. The carbide method worked well for the neptunium isotopes, although the uranium carrier was needed (as it was for the curium). In all of this work, we were greatly aided by the advice and help of the Chemistry Department of the Radiation Laboratory. The beam collection part of the problem turned out to be surprisingly simple. Apparently full efficiency can be achieved using flamed platinum foils. The counting was exclusively by alpha decay, except for the neptunium isotopes. However, the signal-to-noise problem is very serious because of the large number of levels involved. The next slide (Fig. 19) is the Breit-Rabi diagram of the hyperfine levels of the $J = 11/2$ state of neptunium-238, whose nuclear spin is 2. There are 60 levels, but states of angular momenta 9/2, 7/2, and 5/2 are also excited, making a total of 180 magnetic levels in the beam. There are a large number of observable resonances, the intensity of some of which are as low as .07% for some of these heavy elements. This requires tedious counting and many data points for good statistics. Pulse height analysis and precise decay rate was fortunately not needed and would have been very difficult with these intensities, except in the case of neptunium-239, where there was the possibility of a misassignment because of conflict with earlier results.

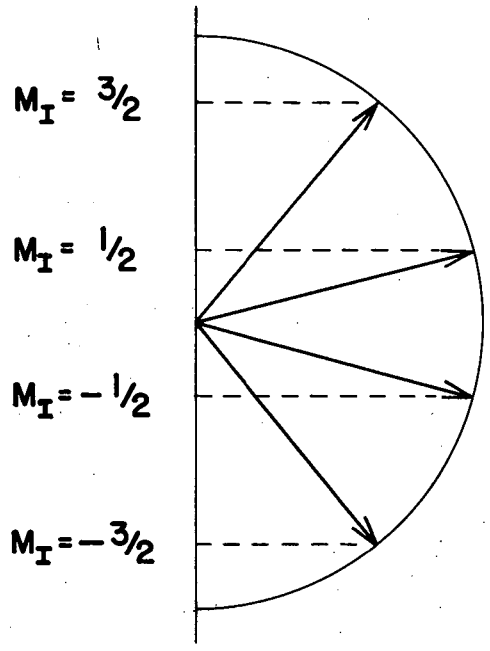
The next slide (Fig. 20) is a summary of the information obtained on the ground state and near ground state configurations for the heavy elements

through curium. The underlined data is from optical measurements, the remainder from atomic beam measurements. The data show that the 5f and 6d electrons are coupled quite differently from the existing assumptions about the rare earths. It is found, to a rather high degree of approximation, that the 5f and 6d shells are quite independent, i.e. that the electrostatic forces on the electrons are small compared to the magnetic forces in the 5f shell. Hence the starting approximation is one where the 5f electrons couple in Russel-Saunders coupling according to Hund's rule to give certain J and g_J values and the 6d electron does the same. These two angular momenta combine according to the usual rules giving final J and g_J values. For curium there are four such states, as previously remarked, and they lie within a few thousand wavenumbers from the ground state and are all found in the atomic beam. From the intensity of the resonances, a reasonable level scheme can be constructed as shown in the next slide (Fig. 21). The four observed g-values can be computed within the observed error by two empirical constants, the g-value of 2.001 for an $^8S_{7/2}$ term for the 5f electrons in combination with the g-value 0.892 for a $^2D_{3/2}$ for the 6d electron, and the calculation is summarized in this slide (Fig. 21). The value 2 for the f electrons represents close to a pure Russel-Saunders situation, the value for the 6d electron is approximately what is expected, but shows a definite discrepancy indicating perturbations. Similar results hold for protactinium, where three of the four possible J states have been seen, for neptunium, where only one level has been seen, and for plutonium, where there are no 6d electrons and the six 5f electrons yield J and g_J values expected from a Russel-Saunders coupling

scheme.

CONCLUSION

It is our aim to continue all these researches, to expand the information on nuclear static moments and to more clearly define the electronic states of the heavy elements. In addition, we are embarked on precision research in these fields for studying hyperfine anomalies and possibly higher nuclear moments. The possibility is very strong in the heavy elements where Z is large and the large I and J values do not generally offer selection rule limitations.



SPACE QUANTIZATION OF SPIN 3/2

MU-16165

Fig. 1. The spatial orientations of a quantized spin 3/2.

RESONANT FREQUENCY FOR SPIN I

WITH $I_0 = 5/2$ RESONANCE AT 10 mc.

$$\nu = \frac{(2I_0 + 1)\nu_0}{(2I + 1)} = \frac{60}{(2I + 1)}$$

I	ν
1/2	30.00
3/2	15.00
5/2	10.00
7/2	7.50
9/2	6.00
11/2	5.00
13/2	4.29

I	ν
1	20.00
2	12.00
3	8.57
4	6.67
5	5.45
6	4.62
7	4.00

MU-11489

Fig. 2. A table of resonant frequencies for an alkali atom where the calibrating isotope is spin 5/2 and resonates at 10 megacycles/second.

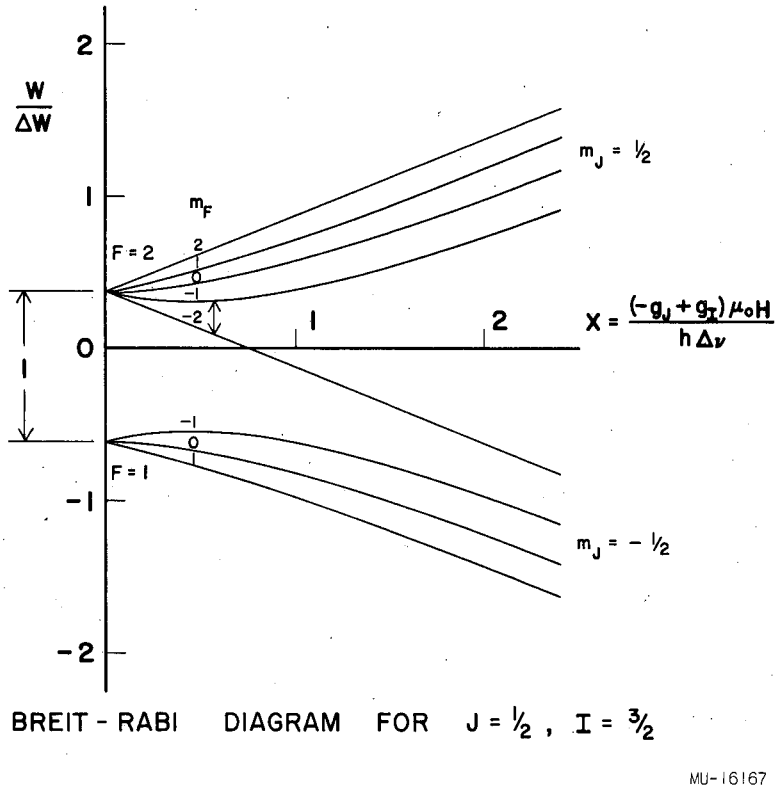


Fig. 3. Breit-Rabi diagram for J of 1/2 and an I of 3/2. Dimensionless energy is plotted versus dimensionless magnetic field.

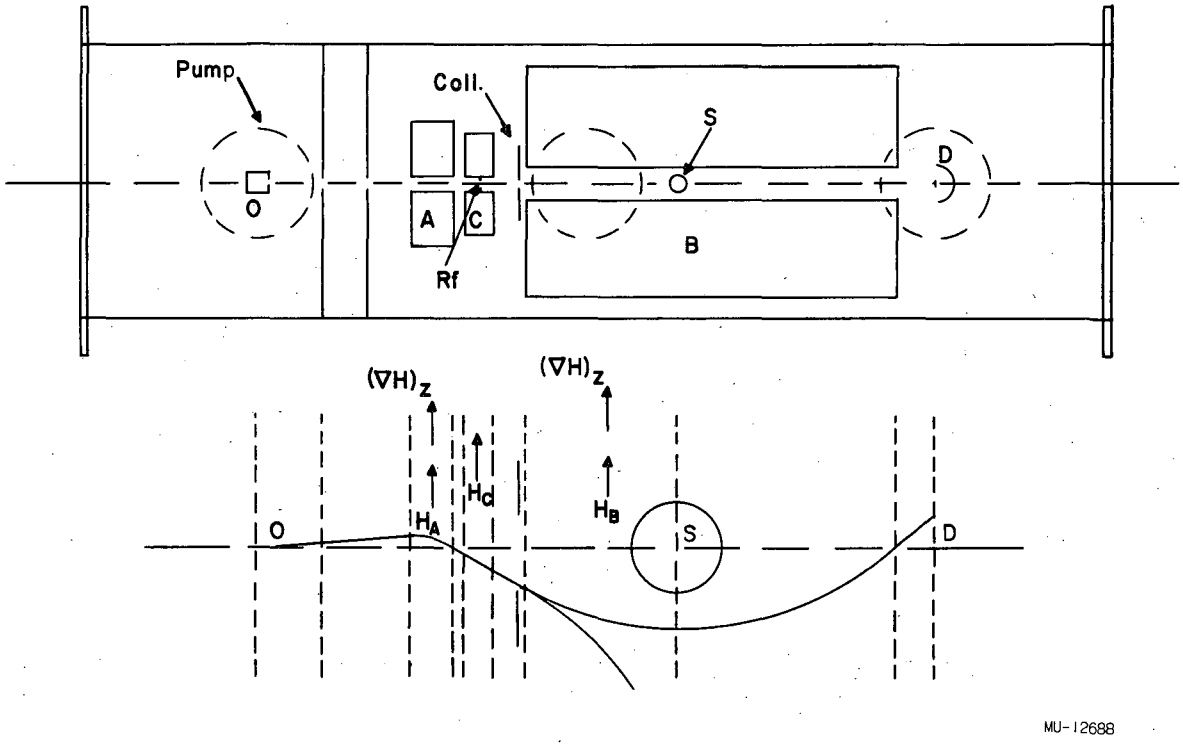
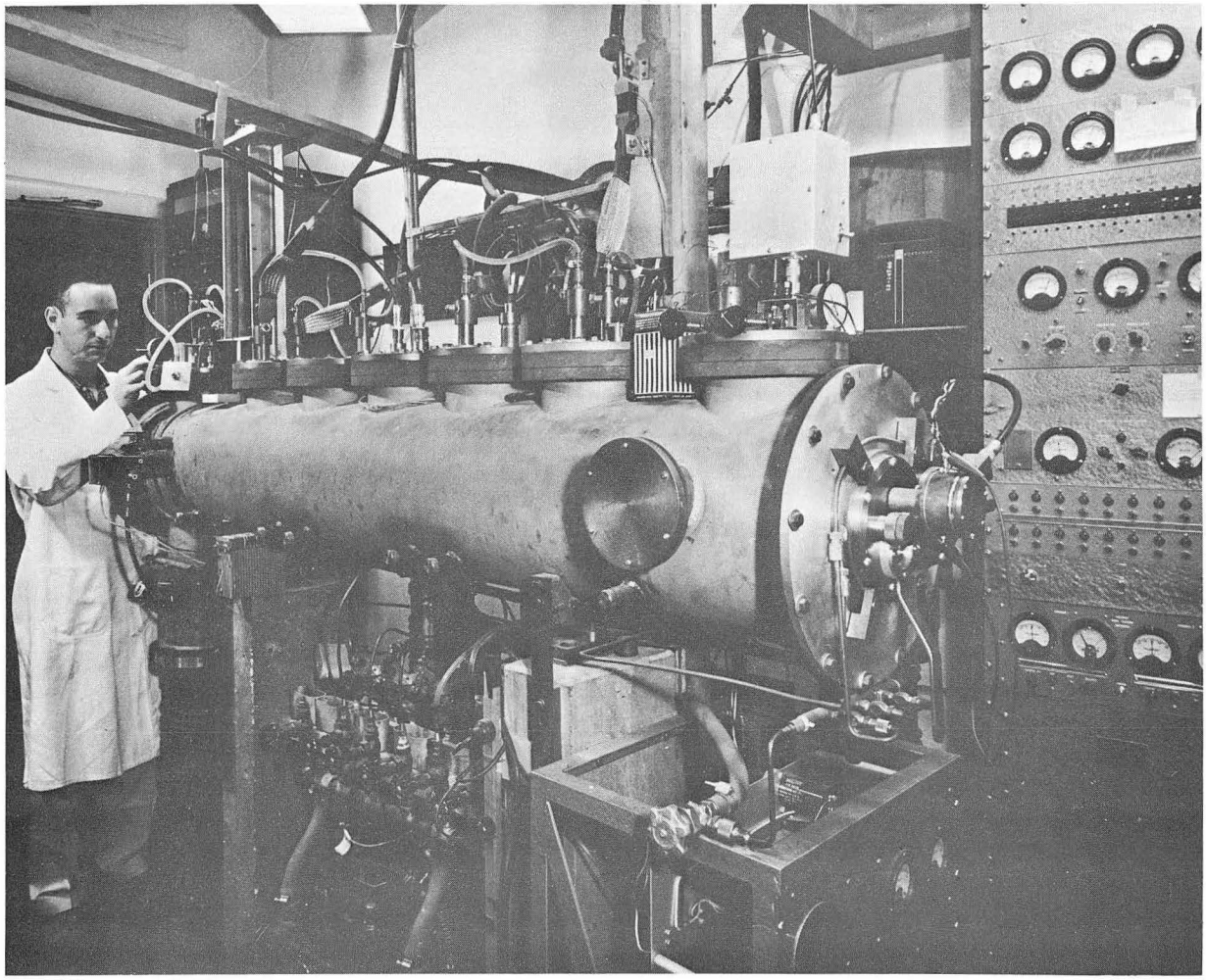


Fig. 4. Schematic sketch of an atomic beam apparatus.



ZN-2055

Fig. 5. Photograph of an atomic beam apparatus.

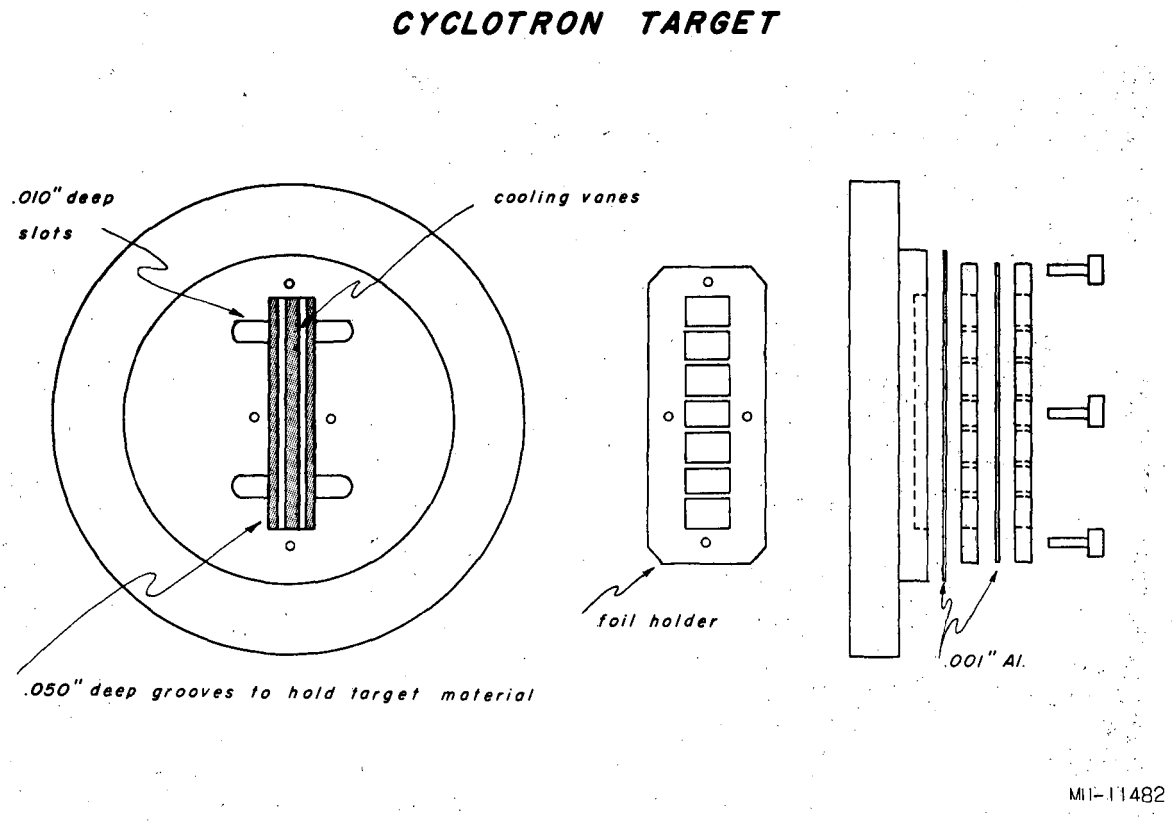


Fig. 6. Exploded view of a cyclotron target.

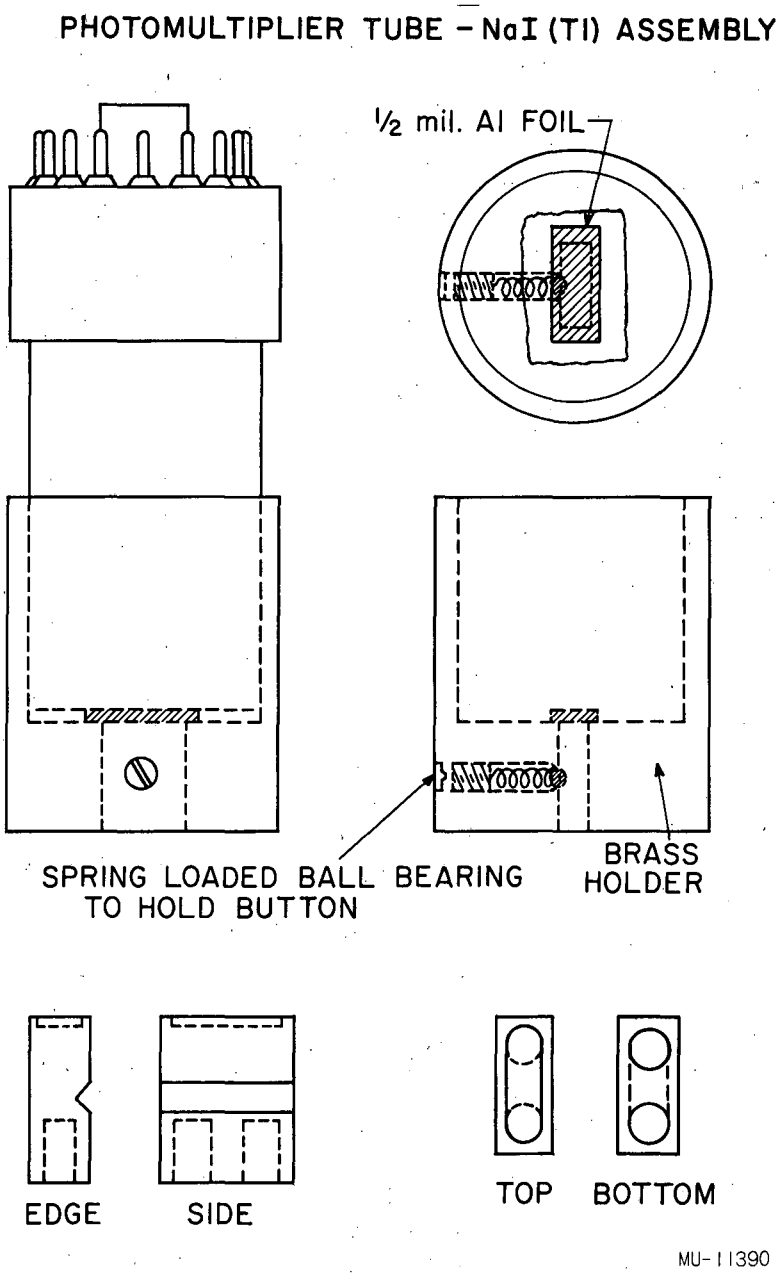


Fig. 7. X-ray counters and collecting button.

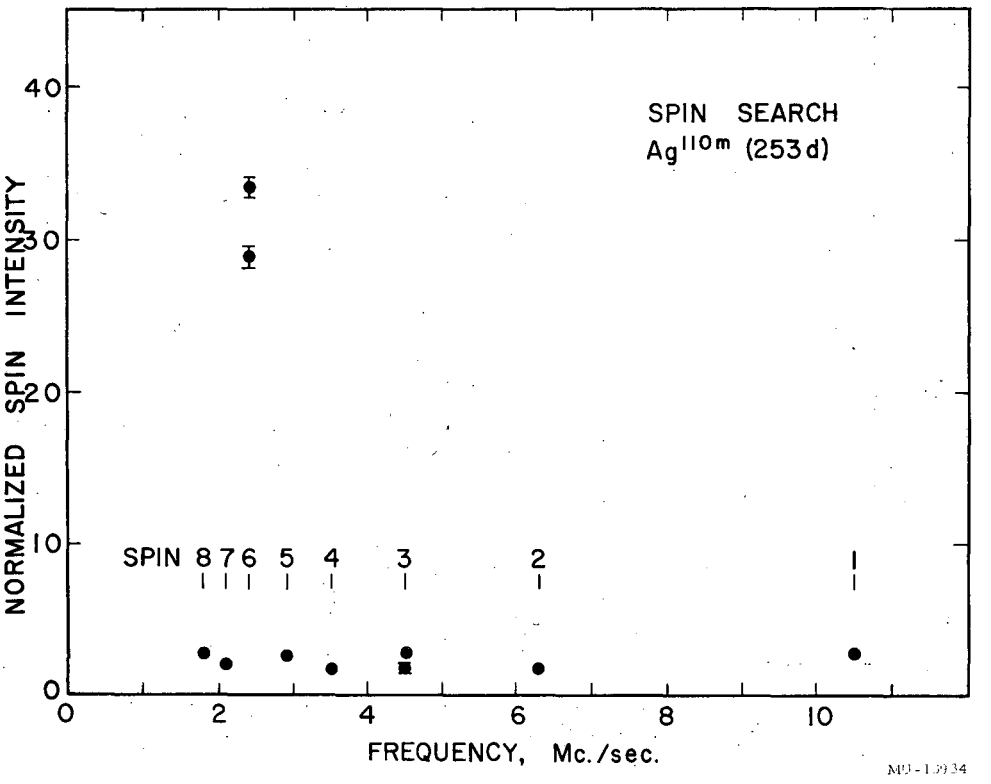


Fig. 8. Spin search for Ag^{110m}. The ordinate is the counting rate, the abscissa is exposure frequency.

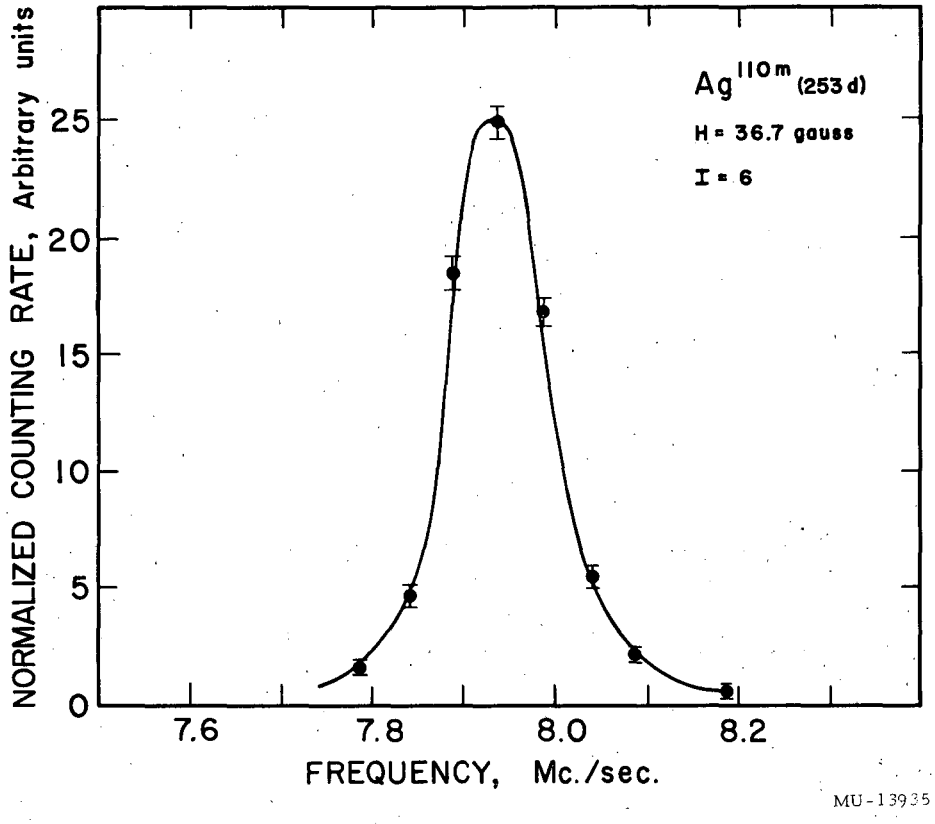


Fig. 9. Resonance line shape of Ag^{110m}.

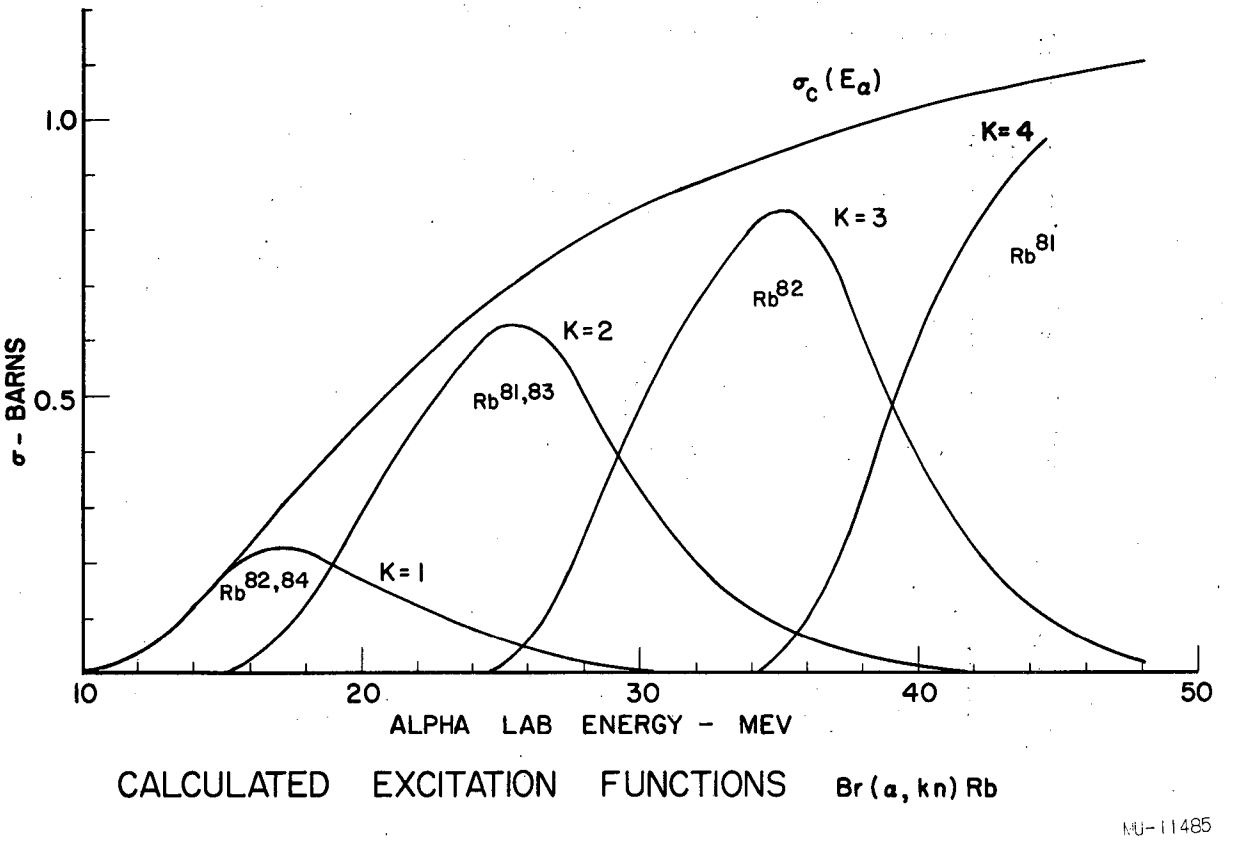


Fig. 10. Production curve for rubidium isotopes as a function of the alpha-particle energy.

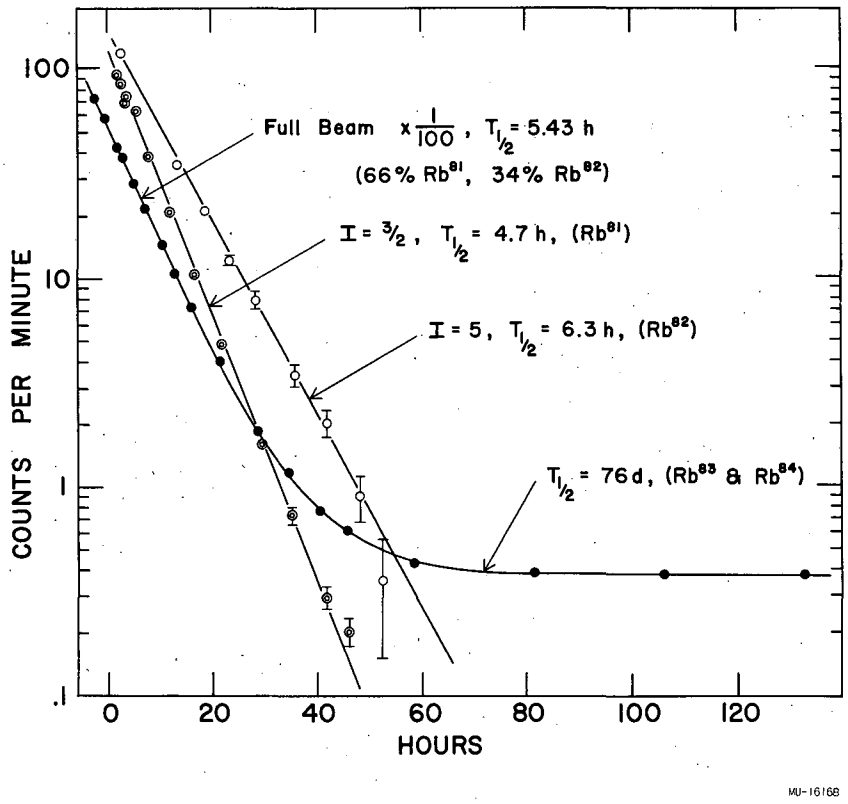


Fig. 11. The decay curves of the Rb^{81} and Rb^{82} resonances compared to the normal beam sample.

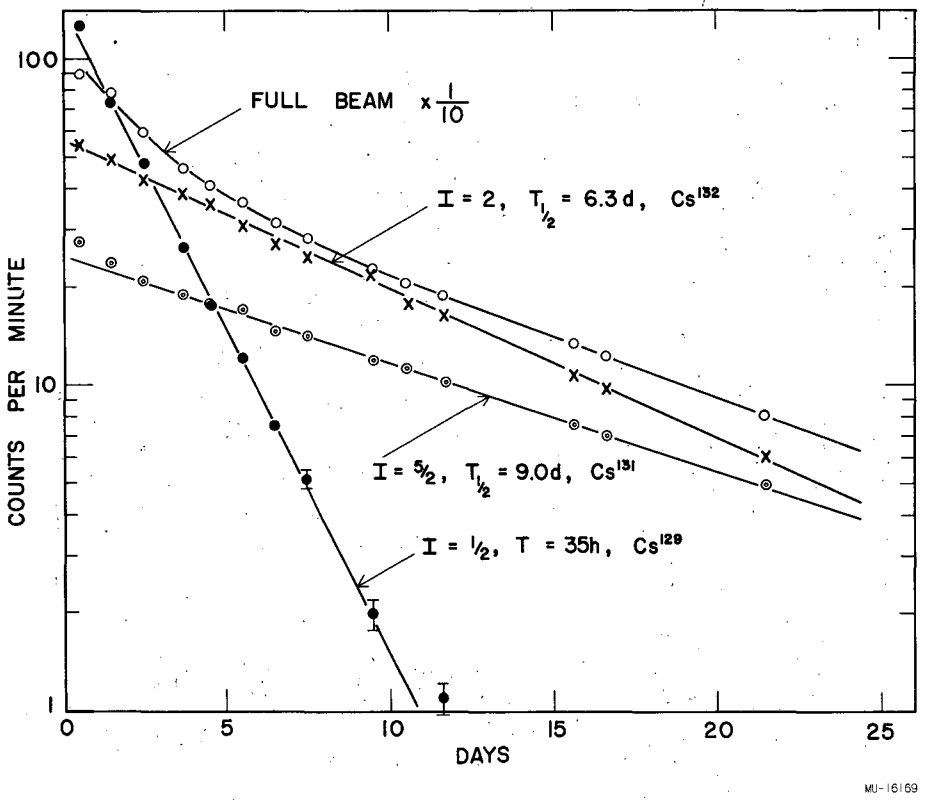


Fig. 12. The decay curves of the Cs^{129}, Cs^{131}, and Cs^{132} resonances compared to the normal beam sample.

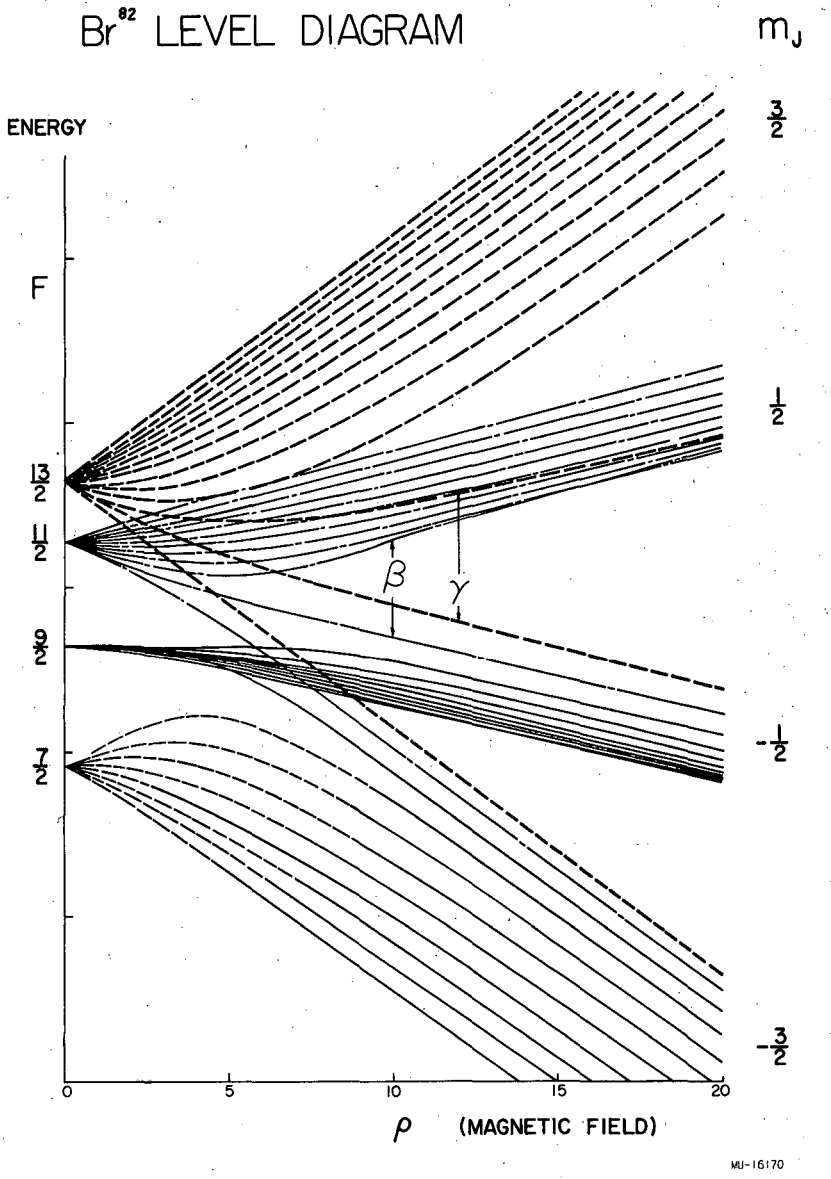


Fig. 13. Breit-Rabi diagram for J = 3/2 and I = 5.

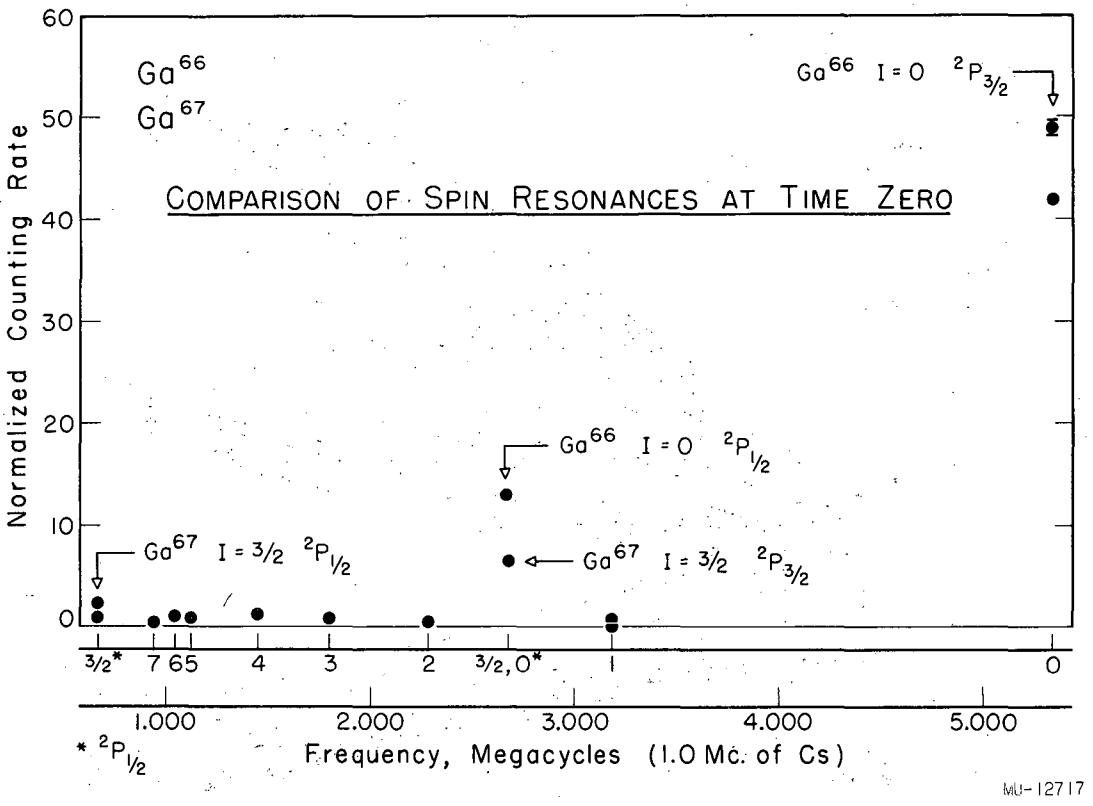


Fig. 14. Spin search involving Ga^{66} and Ga^{67}.

	$T_{1/2}$	Spin	HFS Mc/sec (J=1/2)
$_{19}K^{39}$	stable	3/2	461.71971 (15)
K^{40}	10^9 y	4	1285.790 (7)
K^{41}	stable	3/2	254.018 (6)
K^{42}	12.5 h	2	1258.9 (1)
-K^{43}	22 h	3/2	192.6 (2)
*K^{43}			
$_{29}Cu^{60}$	25 m	2	
Cu^{61}	3.3 h	3/2	
*Cu^{63}	stable	3/2	11,733.83 (1)
Cu^{64}	12.8 h	1	+1282.5 (7)
			-1282.9 (7)
Cu^{65}	stable	3/2	12,568.81 (1)
$_{31}Ga^{66}$	9.4 h	0	
*Ga^{67}	78 h	3/2	
*Ga^{68}	68 m	1	
*Ga^{69}	stable	3/2	
Ga^{71}	stable	3/2	
Ga^{72}	14.1 h	3	
$_{35}$*Br^{76}	17 h	1	
Br^{79}	stable	3/2	
*Br^{80}	18 m	1	
*Br^{80m}	4.5 h	5	
*Br^{81}	stable	3/2	
Br^{82}	35 h	5	
*Br			

* Berkeley measurements
\- Berkeley confirmations

	$T_{1/2}$	Spin	HFS Mc/sec (J=1/2)
$_{37}$*Rb^{81}	4.7 h	3/2	5097 (13)
*Rb^{81m}	31.5 m	9/2	
*Rb^{82}	6.3 h	5	3094.1 (24)
*Rb^{83}	83 d	5/2	3183.3 (58)
*Rb^{84}	33 d	2	3077.5 (51)
Rb^{85}	stable	5/2	3035.735 (2)
Rb^{86}	18.6 d	2	*3948.7 (20)
Rb^{87}	stable	3/2	6834.685 (2)
$_{47}$*Ag^{103}	59 m	7/2	
Ag^{104}?	1.2 h	2	
Ag^{104}	27 m	2	
*Ag^{105}	40 d	1/2	
*Ag^{106}	8.6 d	6	
Ag^{106}	24 m	1	
Ag^{107}	stable	1/2	1712.56 (4)
Ag^{109}	stable	1/2	1976.94 (4)
*Ag^{110m}	253 d	6	
Ag^{111}	7.5 d	1/2	2204.54 (5)
$_{49}$*In^{109}	4.3 h	9/2	
*In^{110m}	5.0 h	7	
*In^{111}	2.8 d	9/2	
In^{113}	stable	9/2	11,385.4300 (20)
In^{113m}	1.73 h	1/2	781.092 (30)
In^{114m}	49 d	5	9700 (200)
In^{115}	stable	9/2	11,409.7506 (20)
In^{116m}	54 m	5	8670 (170)

Berkeley, 10/24/58

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Fig. 15. First summary of results. The asterisks represent Berkeley determinations, the dashes represent Berkeley verifications.

	$T_{1/2}$	Spin	HFS Mc/sec (J=1/2)
53	*I^{123}	13 h	5/2	
	*I^{124}	4.5 d	2	
	I^{125}	60 d	5/2	
	I^{127}	stable	5/2	
	I^{128}	25 m	1	
	I^{129}	1.7×10^7 y	7/2	
	*I^{130}	12.6 h	5	
	-I^{131}	8 d	7/2	
55	*Cs^{127}	6.2 h	1/2	8950 (200)
	*Cs^{129}	31 h	1/2	9200 (200)
	*Cs^{130}	30 m	1	+6400 (350)
				-6800 (350)
	-Cs^{131}	10 d	5/2	13,200 (110)
	*Cs^{132}	6.2 d	2	8648 (35)
	Cs^{133}	stable	7/2	9192.631770 (20)
	Cs^{134}	2.3 y	4	10,473.626 (15)
	Cs^{134m}	3.1 h	8	3684.594 (20)
	Cs^{135}	10^6 y	7/2	9724.023 (15)
	Cs^{137}	30 y	7/2	10,115.527 (15)
79	*Au^{191}	3 h	3/2	6000 (200)
	*Au^{192}	4.8 h	1	374.3 (2)
	*Au^{193}	17 h	3/2	6000 (200)
	-*Au^{194}	39 h	1	3600 (120)
	*Au^{195}	185 d	3/2	
	*Au^{196}	5.6 d	2	
	Au^{197}	stable	3/2	6107.1 (10)
	Au^{198}	2.7 d	2	+21,800 (150)
				-22,500 (150)
	Au^{199}	3.2 d	3/2	11,180 (130)

* Berkeley measurements
\- Berkeley confirmations

	$T_{1/2}$	Spin	HFS Mc/sec (J=1/2)
81	*Tl^{197}	2.8 h	1/2	
	*Tl^{198m}	1.9 h	7	
	*Tl^{199}	7.4 h	1/2	
	*Tl^{200}	27 h	2	
	*Tl^{201}	3 d	1/2	
	*Tl^{202}	12 d	2	
	Tl^{203}	stable	1/2	21,105.447 (5)
	*Tl^{204}	4 y	2	732 (5)
	Tl^{205}	stable	1/2	21,310.835 (5)
83	Bi^{203}	12 h	9/2	
	Bi^{204}	12 h	6	
	Bi^{205}	14 d	9/2	
	*Bi^{206}	6.4 d	6	
	Bi^{209}	stable	9/2	
	Bi^{210}	10^6 y	1	
85	*At^{211}	7.2 h	9/2	
91	Pa^{231}	34,000 y	3/2	
	*Pa^{233}	27.4 d	3/2	
93	Np^{237}	2×10^6 y	5/2	
	*Np^{238}	2.1 d	2	
	*Np^{239}	2.3 d	5/2	
94	-Pu^{239}	24,000 y	1/2	*7.683 (60)
95	-Am^{241}	461 y	5/2	
	Am^{243}	8000 y	5/2	
96	*Cm^{242}	165 d	0	

Berkeley 10/24/58

MU-16172

Fig. 16. Second summary of results.

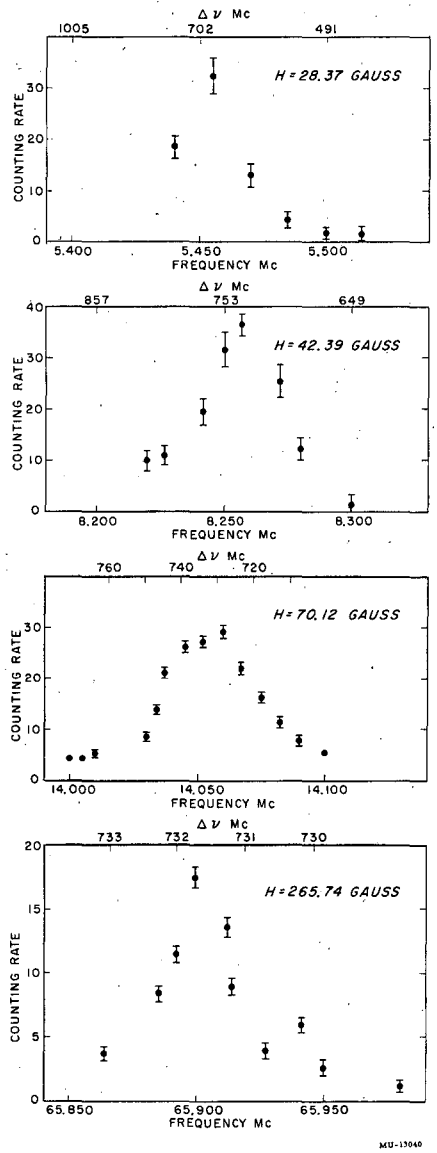

Fig. 17. Tl^{204} resonances as a function of increasing magnetic field. The scale of equivalent hyperfine separations is at the top of each graph.

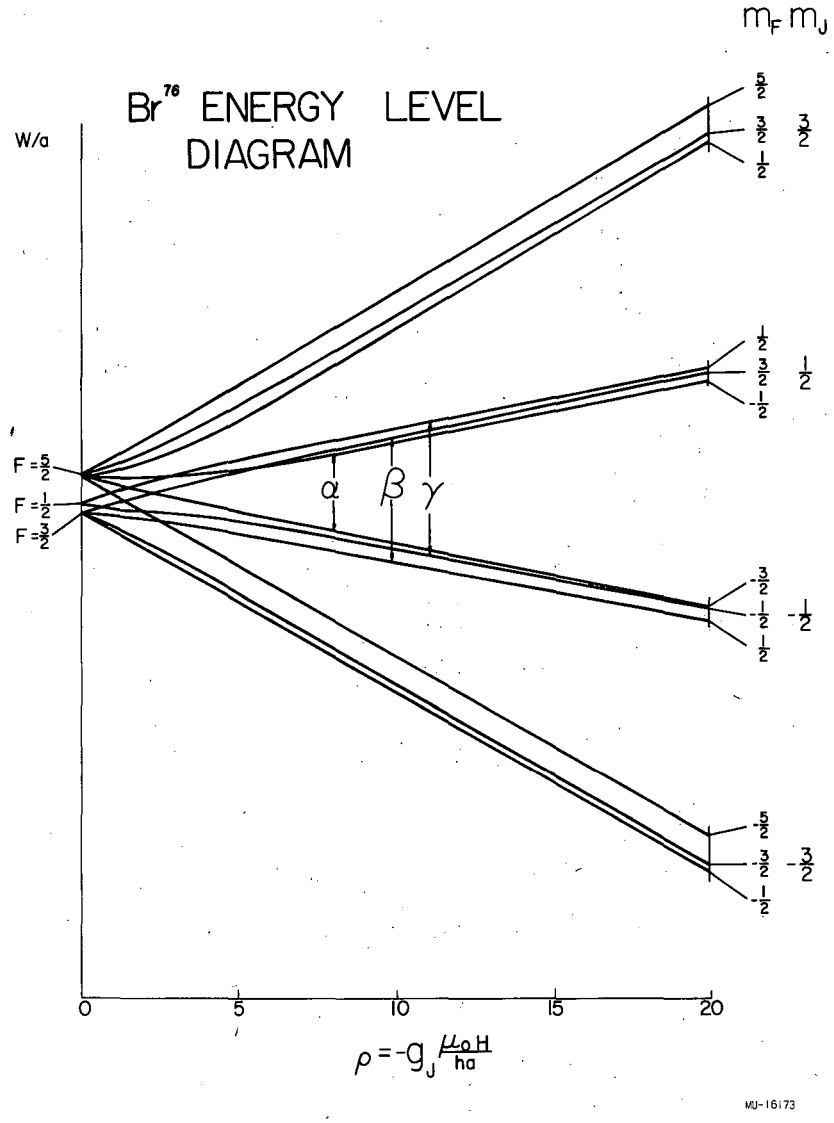

Fig. 18. Breit-Rabi diagram for J = 3/2, I = 1, inverted as for Br^{76}.

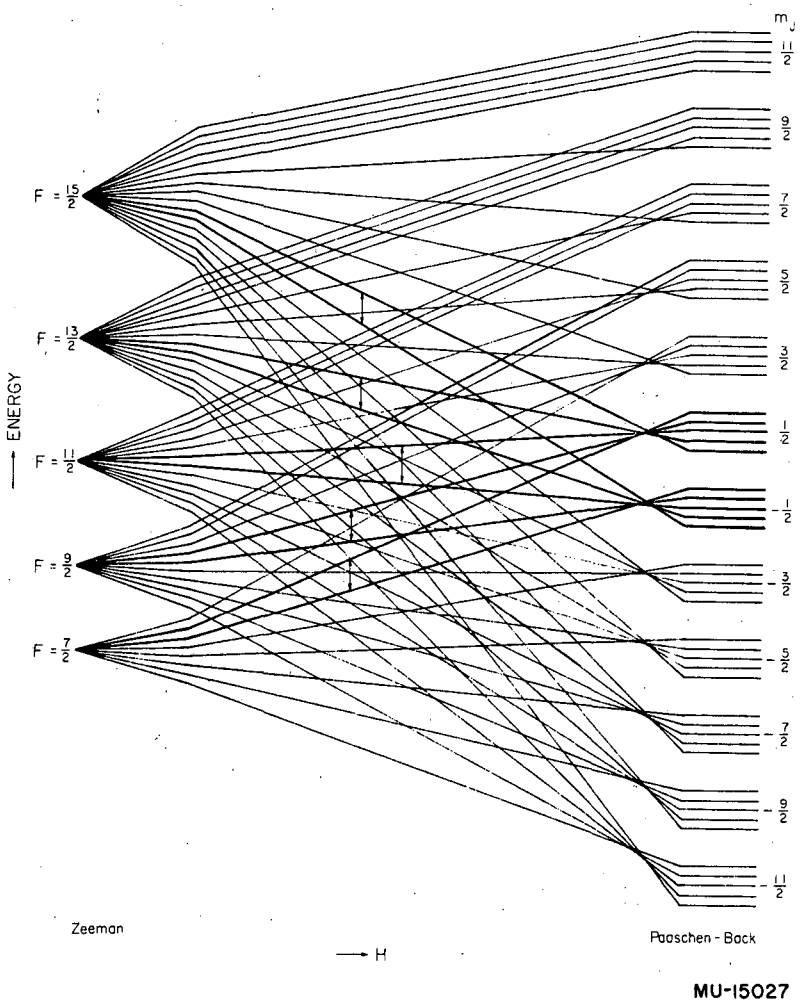

Fig. 19. Breit-Rabi diagram for J = 11/2, I = 2 as for neptunium-238.

HEAVY ELEMENT GROUND STATES

ELEMENT	CONFIGURATION	GROUND STATES	SPINS
^{89}Ac	$6d^1(7S)^2$	$J=\frac{3}{2}$	
^{90}Th	$(6d)^2\ (7S)^2$	$J=2$ $\quad g_J=.74$	
^{91}Pa	$(5f)^2\ (6d)^1\ (7S)^2$	$J=\frac{7}{2}$ $\quad g_J=.7922$ (30) $J=\frac{9}{2}$ $\quad g_J=.8066$ (10) $J=\frac{11}{2}$ $\quad g_J=.8144$ (10)	Pa^{233} $I=\frac{3}{2}$
^{92}U	$(5f)^3(6d)^1(7S)^2$	$J=6$ $\quad g_J=.73$	
^{93}Np	$(5f)^4(6d)^1\ (7S)^2$	$J=\frac{11}{2}$ $\quad g_J=.6551$ (6)	Np^{238} $I=2$ Np^{239} $I=\frac{5}{2}$
^{94}Pu	$(5f)^6\ (7S)^2$	$J=1$ $\quad g_J=1.4975$ (10)	Pu^{239} $I=\frac{1}{2}$
^{95}Am	$(5f)^7\ (7S)^2$	$J=\frac{7}{2}$ $\quad g_J\approx 2$	Am^{241} $I=\frac{5}{2}$
^{96}Cm	$(5f)^7\ (6d)^1\ (7S)^2$	$J=5$ $\quad g_J=1.6705$ (27) $J=4$ $\quad g_J=1.7760$ (15) $J=3$ $\quad g_J=2.0000$ (25) $J=2$ $\quad g_J=2.5613$ (27)	Cm^{242} $I=0$

MU-16174

Fig. 20. Summary of information on the low-lying states of the heavy elements. The underlined data are from optical measurements, the remainder from atomic beam work.

ELECTRONIC COUPLING IN CURIUM

The independence of 5f and 6d electrons, plus pure L-S coupling in the 5f shell would predict:

A. 7 5f electrons $S=\frac{7}{2}$ $L=0$ $^8S_{\frac{7}{2}}$ $g_J = 2.002$

B. 1 6d electron $S=\frac{1}{2}$ $L=2$ $^2D_{\frac{3}{2}}$ $g = .800$

These angular moments would then couple weakly to give 4 close-lying J-states, J = 2,3,4,5.

Experimental Data

Observe	J = 2	$g_J = 2.5613$ (27)	GROUND STATE
	J = 3	$g_J = 2.0000$ (25)	550 Cm^{-1}
	J = 4	$g_J = 1.7760$ (15)	1200 Cm^{-1}
	J = 5	$g_J = 1.6705$ (27)	2200 Cm^{-1}

These observations are fit to the experimental accuracy by:

$J_1 = \frac{7}{2}$ $g_{J_1} = 2.001$

$J_2 = \frac{3}{2}$ $g_{J_2} = .89$

Conclusion

A. Pure L-S coupling is a good approximation for 5f electrons.

B. 5f & 6d electrons are weakly coupled.

C. 6d electrons appear to be mixed with higher configurations.

MU-16166

Fig. 21. The g-factors for the electronic subshells of the low-lying states of curium.