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HYPERFINE STRUCTURE AND NUCLEAR MOMENTS OF PROMETHIUM-147 AND PROMETHIUM-151

Berkeley, California

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ABSTRACT

The atomic-beam magnetic-resonance method has been used to measure the hyperfine structure of the $^6H_{7/2}$ level in two promethium isotopes. The spins of these isotopes have been verified to be $I(Pm^{147}) = 7/2$ and $I(Pm^{151}) = 5/2$. The electronic splitting factor (g_J) has been measured for the $^6H_{7/2}$ level and is found to be $g_J = -0.8279(4)$, in good agreement with the predicted value. The hyperfine constants and the nuclear moments inferred from them are found to be

for Pm^{147}:	$\lvert A\rvert = 447.1(9.3)$ Mc	$\lvert B\rvert = 267.5(70.8)$ Mc	$B/A < 0$
	$\lvert\mu_I\rvert = 3.2(3)$ nm	$\lvert Q\rvert = 0.7(3)$ b	$Q/\mu > 0$;

and

for Pm^{151}:	$\lvert A\rvert = 358(23)$ Mc	$\lvert B\rvert = 777(94)$ Mc	$B/A < 0$
	$\lvert\mu_I\rvert = 1.8(2)$ nm	$\lvert Q\rvert = 1.9(3)$ b	$Q/\mu > 0$.

The nuclear moments are corrected for the breakdown of Russell-Saunders coupling. The stated errors include uncertainties in the fields at the nucleus arising from errors in the value of $\langle 1/r^3 \rangle$ and the neglect of core polarization on the magnetic hyperfine structure. Corrections due to the Sternheimer effect have not been included. The measurements are shown to be consistent

with the hypothesis that Pm^{147} is not highly deformed and can be understood from the shell model, but that Pm^{151} is highly deformed and must be interpreted by the collective model.

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INTRODUCTION

It is an experimentally well-established fact that collective effects become important in nuclei for neutron numbers N greater than about 88. The consequences of collective motion for the nuclear moments of a pair of isotopes can be very striking. An example, pertinent to our experiment, is the moments of Eu^{151} and Eu^{153} with neutron numbers 88 and 90, respectively. For Eu^{153}, which is highly deformed, the spectroscopic quadrupole moment is about twice that of Eu^{151}. Moreover, the measured magnetic moment of Eu^{153} is considerably less than that of Eu^{151}, and is one of the rare examples of a magnetic moment that lies in the wrong Schmidt group.[1]

For the pair of promethium isotopes under investigation here, the neutron numbers are 86 and 90. Part of the theoretical incentive for this work was to attempt to establish the onset of collective effects for N greater than 88 as valid for the isotopes of promethium. That our results are strikingly similar to those found in europium will be seen shortly.

Earlier work on these isotopes had already established the spins and parities of the nuclear ground states. The spin of Pm^{147} had been shown[2] to be 7/2 and that of Pm^{151} to be 5/2.[3] If the nuclear core is unmodified by the addition of neutron pairs, the spins of a set of odd-Z isotopes should remain unchanged. Hence, the change in spin can be interpreted as evidence for the onset of collective motion.

Many workers have investigated the beta decay from the ground state of promethium-147 and have assigned positive parity to this state. The population of the energy levels of Pm^{151} by the beta decay of Sm^{151} has been investigated by Schmid and Burson.[4] They assign positive parity to the Pm^{151} ground state. In addition, they find evidence for the existence of a rotational band among the observed levels if it is assumed that the ground state is characterized either by $K = 1/2$ or $9/2$. In view of the subsequently measured spin and the energy-level scheme of Mottelson and Nilsson, this conclusion seems dubious.[5] Chéry has also assigned the parity of the promethium-151 ground state as positive on the basis of an extensive investigation of the beta decay from the ground state.[6] In our subsequent discussion of the nuclear moments of Pm^{151}, we will assume the positive parity assignment by these workers.

Further theoretical incentive for this work comes from the recent calculations by Judd and Lindgren for the electronic splitting factors g_J of the low-lying levels that arise from the configuration $(4f)^n$ in the rare earths.[7] Including spin-orbit, relativistic, and diamagnetic corrections, they obtain for the $J = 7/2$ level of the promethium ground term $g_J = -0.8275$. A precise determination of the g_J value of this state will, therefore, serve as a check on the parameters they use to characterize the electronic state.

BEAM PRODUCTION AND DETECTION

Promethium-147 is a fission product and can be readily purchased in the form of the chloride in curie quantities.[8] A spectroscopic analysis and a pulse-height spectrum showed that some shipments of Pm were contaminated

with americium-241 and isotopes of samarium. However, neither of these could confuse the Pm results.

The chemistry used to produce a beam of atomic promethium is straightforward. An excess of nitric acid is added to 1 curie of the chloride and the nitrate product is reduced to about 1 ml. This is pipetted into a sharp-edged tantalum crucible and slowly heated to 700° F to form the oxide. To convert the oxide to the metal, a reducing agent is added and the crucible is heated in the atomic-beam oven. In this experiment both misch metal and carbon were tried, each with partial success; in the later stages of the work misch metal was used exclusively. A really satisfactory beam was never achieved, and in order to obtain a signal-to-noise ratio of 1:1 it is necessary to employ beam intensities at the detector of the order of 5000 counts/min.

Promethium-151 is produced by neutron irradiation of natural neodymium metal. The material is bombarded for 4 days at a flux of 9×10^{13} at the GE reactor in Vallecitos. This bombardment yields approximately equal curie amounts of Pm^{151}, Pm^{149}, and Nd^{147} and gives rise to a large background from the undesired isotopes. Another source of background comes from atoms in the $J = 5/2$ ground electronic level, so that the signal-to-noise ratio for Pm^{151} is no better than that for Pm^{147}. Unfortunately, the electronic moment of atoms in the $J = 5/2$ level is too small for the magnets to deflect them around the stop wire. They are therefore useless for hyperfine-structure measurements.

Promethium beams are detected by collection on freshly flamed platinum foils which are counted in Geiger counters. The collection efficiency of platinum for promethium is greater than 25 % and is highly reproducible.

EXPERIMENTAL METHOD AND OBSERVATIONS

Our measurements were all performed by use of the atomic-beam method of Zacharias.[9] In this scheme, transitions are observed between hyperfine levels whose high-field magnetic moments are equal and opposite and for which $\Delta F = 0, \pm 1$; $\Delta M_F = 0, \pm 1$. In Fig. 1, schematics are shown for the hyperfine structures of Pm^{147} $(I = J = 7/2)$ and Pm^{151} $(J = 7/2, I = 5/2)$. It is seen that for both isotopes there are four transitions of the type $\Delta F = 0$, $\Delta M_F = \pm 1$ that satisfy the high-field selection rule as well. The transition in the highest F states is labeled α, the next β, and so on. These transitions were each observed at several values of the magnetic field. Sample resonances for Pm^{147} are shown in Figs. 2 and 3, and for Pm^{151} in Fig. 4.

The observed field-dependent data can be analyzed by a Hamiltonian of the form

$$\mathcal{H} = A\,\vec{I}\cdot\vec{J} + \frac{B}{2IJ(2I-1)(2J-1)}\left[3(\vec{I}\cdot\vec{J})^2 + 3/2(\vec{I}\cdot\vec{J}) - I(I+1)J(J+1)\right] - g_J\mu_0\,\vec{J}\cdot\vec{H}\,, \tag{1}$$

where A and B are the magnetic-dipole and electric-quadrupole hyperfine constants, respectively. In this analysis, the quantities A, B, and g_J are treated as parameters, and a least-square fit to the data is obtained. The method is described elsewhere.[10] The values obtained for the data are

for Pm^{147}: $|A| = 447(9)$ Mc, $|B| = 267(71)$ Mc, and $g_J = -0.8283(4)$;

for Pm^{151}: $|A| = 358(22)$ Mc, $|B| = 778(93)$ Mc, and $g_J = -0.8272(7)$.

From these we obtain $g_J = -0.8279(4)$ as the weighted mean.

With these values of the hyperfine constants, the theoretical frequencies have been calculated from the Hamiltonian (1). These are compared with the measured frequencies in Tables I and II. The stated errors in the quantities represent standard deviations.

The promethium-147 used in these experiments is guaranteed by the supplier to be the stated isotope. There are, however, independent checks on its identity. That there is Pm^{147} present is shown by the measured g_J and J values. Of the promethium isotopes that are fission products only one (Pm^{147}) has a half-life greater than 53 h. The half-life of our sample is known to be much greater than 53 h. The promethium-151 is identified on the basis of the method of production and the measured half-life.

CALCULATION OF THE NUCLEAR MOMENTS

In order to infer the nuclear moments from the measured hyperfine structure it is necessary to make some assumptions about the electronic structure of the promethium ground state. As a first approximation, it is assumed that the level under consideration arises from pure Russell-Saunders coupling among the electrons of the configuration $(4f)^5$ to the Hund's-rule term $^6H_{7/2}$. Strong support for this assumption comes from the small discrepancy between the measured g_J value of -0.8279 and the g_J value of -0.8250 predicted from pure Russell-Saunders coupling. The expressions for calculating the magnetic-dipole and electric-quadrupole fields at the nucleus for the Hund's-rule term have been given elsewhere.[11] We state here the results

$$\langle H_Z \rangle = -7.49\, \mu_0 \langle 1/r^3 \rangle_{4f}$$

and (2)

$$\langle q_J \rangle = \frac{26}{75} \langle 1/r^3 \rangle_{4f} .$$

The expressions used for the fields at the nucleus are such that the hyperfine coupling constants are given by $A = -\frac{1}{IJ} \langle H_Z \rangle \mu_I$, $B = -e^2 \langle q_J \rangle Q$.

The question of the appropriate value of $\langle 1/r^3 \rangle$ for 4f electrons has received considerable attention in the literature. An early attempt was made by Bleaney for the triply ionized rare earths.[12] Bleaney showed that a hydrogenic expression for the spin-orbit coupling energy could reasonably fit the measured energies. These were then used to calculate values for $\langle 1/r^3 \rangle$. More recently Judd and Lindgren have developed a set of modified hydrogenic functions.[7] The parameters are chosen to give agreement with self-consistent field calculations for Pr^{3+} and Tm^{3+}, and with the spin-orbit splitting constants of several of the rare earth atoms. The values obtained in this way for triply ionized atoms are found to be about 25 % smaller than those of Bleaney. Lindgren has used these values to redetermine all rare earth moments inferred from hyperfine-structure work. The latest attempts at $\langle 1/r^3 \rangle$ calculations are those of Freeman and Watson.[13] These authors have performed Hartree calculations for rare earth ions, and found that their values differ considerably from those of Judd and Lindgren and are within 5 % of Bleaney's. They note that in principle the Hartree $\langle 1/r^3 \rangle$'s are more suited to evaluating nuclear-moment data. This follows from the fact that an effective $\langle 1/r^3 \rangle$ is used in determining the modified hydrogenic function, whereas the actual $\langle 1/r^3 \rangle$ comes into the nuclear-moment calculation.

Recently, rare-earth magnetic moments have been directly measured for Nd^{143}, Er^{169}, Tm^{169}, and Yb^{171}. The values obtained for erbium, thulium, and ytterbium all lie within 5 % of the moments inferred from hyperfine data, obtained by using the $\langle 1/r^3 \rangle_{4f}$ values of Lindgren. However, the ground configurations of these elements are $(f)^{12}$, $(f)^{13}$, and $(f)^{14}$, respectively, and they constitute the last three elements in the lanthanide series. The ground configuration of Pm is $(f)^5$ and lies closest to Nd, which is $(f)^4$. Recently, Halford[14] has completed a measurement on $(Nd^{143})^{3+}$ and finds a magnetic moment close to the prediction by Freeman and Watson, but it differs from Lindgren's prediction by more than 15 %. This is substantially greater than the 5 % uncertainty that Lindgren assigns to his $\langle 1/r^3 \rangle$'s. The situation at present seems to be that Lindgren's $\langle 1/r^3 \rangle_{4f}$ values work well for the heaviest lanthanides, but they do not give agreement with Nd, which is in the early part of the rare earth group.

In our calculations we use Lindgren's value of $\langle 1/r^3 \rangle$ for promethium and incorporate his estimated uncertainty of 5 % into our error. However, we note here some concern that 5 % may not be a realistic figure. When this is done, we find for the nuclear moments of Pm^{147}: $\mu_I = \pm 3.19$ nm and $Q = \pm 0.7$ b; and for Pm^{151}: $\mu_I = \pm 1.82$ nm and $Q = \pm 1.9$ b.

The value obtained for the nuclear moment of Pm^{147} is in good agreement with a value of +3.0 nm obtained by paramagnetic resonance.[15] There is a discrepancy, however, with a value reported on the basis of optical spectroscopic measurements.[16] More recent measurements on the optical spectrum of promethium-147 lead to the suggestion, however, that the earlier optical spectroscopy results may have been in error.[17]

We now consider possible effects that could give deviations from the values quoted above. There is, first of all, a perturbation due to the spin-orbit interaction which leads to the breakdown of Russell-Saunders coupling. Such a perturbation has the effect of causing the g_J value to deviate from the Russell-Saunders value.

In first-order perturbation theory, the spin-orbit interaction admixes the ground level with the $^4G_{7/2}$ level. There are four possible ^4G terms that can be formed from the configuration $(f)^5$. These may be distinguished from one another with the help of group theoretical methods and the separations calculated.[18] To find the linear combination that corresponds to the lowest eigenvalue, the matrix of the Coulomb interaction within the ^4G terms must be diagonalized. This gives

$$\left|{}^4G_{7/2}\right\rangle = 0.484\left|{}^4G(211)\,(20)\right\rangle + 0.129\left|{}^4G(211)\,(21)\right\rangle - 0.717\left|{}^4G(211)\,(30)\right\rangle - 0.471\left|{}^4G(111)\,(20)\right\rangle .$$

As a first approximation to the electronic wave function, we may let

$$\psi = [1 - \alpha^2]^{1/2}\left|{}^6H_{7/2}\right\rangle + \alpha\left|{}^4G_{7/2}\right\rangle$$

and adjust α to give the measured g_J value. In this way, we find $\alpha = 0.137$. With this value, the correction to the magnetic field at the nucleus is of order α and can be evaluated by spherical tensor methods. It is found that the correction to the magnetic moment is 2%.[19] The correction to the quadrupole field is of order α^2 and is accordingly neglected.

We now consider the core-polarization correction to the calculated moment. This arises from the existence of unparied s electrons due to

exchange interaction with the valence electrons. It has been observed in atoms having half-filled closed shells, e.g., N, Mn, and Am. Bleaney has argued that core-polarization effects in the triply ionized rare earths are small, and estimates that it is about 2.9% for triply ionized promethium.[12] For the free atom, core polarization also includes contributions from a 6s electron pair. We can estimate the 6s effect in praseodymium from the values obtained for a_{4f} in stable and triply ionized Pr^{141}. Let

$$\left(\frac{a_{4f}}{\langle 1/r^3 \rangle}\right)_{Pr^{3+}} \Bigg/ \left(\frac{a_{4f}}{\langle 1/r^3 \rangle}\right)_{Pr} = 1 + \Delta . \tag{3}$$

The a_{4f}'s are experimental quantities derived from the measured hyperfine structure, without including possible core-polarization effects. Hence the quantity Δ will be a measure of the degree of 6s core polarization. For Pr we obtain $\Delta = 1.4\%$. The 6s effect in promethium should not be substantially different, and it is felt that the correction to the moment should not be greater than about 5%. This has been incorporated into the uncertainty.

DISCUSSION

A comparison of the measured moments of Pm^{147} and of Pm^{151} yields two important qualitative points. Firstly, the quadrupole moment of Pm^{151} is considerably larger than that of Pm^{147} and cannot be interpreted by any reasonable single-particle model. Secondly, the magnetic moment of Pm^{151} is considerably smaller than that of Pm^{147} and lies in the wrong Schmidt group. As noted in the introduction, both these effects are observed

in a comparison of the moments of the isotopes Eu^{151} and Eu^{153}. They can be easily explained by the assumption that the nuclear core of the promethium isotopes becomes deformed for Pm^{151}, and are striking confirmation of the hypothesis of a transformation of nuclear shape above $N = 88$.

That a deformed nuclear core can give large quadrupole moments is obvious. That nuclear moments can lie in the wrong Schmidt group for deformed nuclei can be understood by recognizing that the interaction of a deformed core with the odd nucleon is noncentral. Hence the orbital angular momentum, ℓ, is no longer a good quantum number, and can take on all odd or all even values less than or equal to N, the total oscillator quantum number. However, the projection of the total single-particle angular momentum on the axis of nuclear symmetry, Ω, is a good quantum number, and it is equal to $\Lambda + \Sigma$, the sum of the projections on the symmetry axis of the orbital and spin angular momentum. Hence the wave function for a nucleus of given spin and parity in general contains some states with $\Omega = \Lambda + 1/2$ and some with $\Omega = \Lambda' - 1/2$, and can therefore lie in either the Schmidt group or between the Dirac lines. The actual location of the moment on the Schmidt diagram depends on the other parameters that define the state.

Let us now consider the specific possibilities for the state assignments. From the shell model we can predict the spin and parity of Pm^{147} if the odd proton is assigned to the $g_{7/2}$ level. The quoted moment of Pm^{147} lies just above the Dirac line; however, it is within the stated uncertainty of the Schmidt group. The quadrupole moment can be calculated from the assumption of three $J = 7/2$ holes coupled to a spin of $J = 7/2$. Using $\langle r_n^2 \rangle = R_0^2 A^{2/3}$ with $R_0 = 1.2 \times 10^{-13}$ cm, we obtain $Q = +0.09$ b. This is considerably smaller than the measured quadrupole moment, and conceivably is due to configuration

mixing or small deformations of the nuclear core. The only other shell-model state likely to contain the 61 st proton is $d_{5/2}$, but this is not allowed because the configuration $(d_{5/2})^3$ coupling to $I = 7/2$ is forbidden by the Pauli principle.

On the assumption that the nuclear core of Pm^{151} is highly deformed, there are two possible state assignments for the 61 st proton that give the correct spin and parity. When the notation of Mottelson and Nilsson is used,[5] these are $5/2 + [413]$ and $5/2 + [402]$. We have calculated the nuclear moments of these states for different values of the deformation parameter δ. The value obtained for the level $5/2 + [402]$ is about 3.7 nm and is insensitive to the deformation. The level $5/2 + [413]$ gives a moment of 0.91 nm with a deformation parameter of $\delta \approx 0.4$. This is in better agreement with the measured value and seems to us to be the proper state assignment. The collective-model value for the quadrupole moment can be obtained from the expression

$$Q = Q_0 \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}, \tag{4}$$

where $Q = 4/5\ ZR_0^2\delta$. Using these expressions, we obtain for the quadrupole moment $Q = +2.1$b. The theoretical sign of Q/μ is positive, which agrees with the sign inferred from the data.

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FOOTNOTES AND REFERENCES

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Table I. Summary of observations in Pm^{147}.

H (gauss)	Transition	ν(exp) (Mc/sec)	Residuals (Mc/sec)
20.75	α	12.06	+0.009
38.24	α	22.28	+0.035
71.63	α	41.815	+0.006
117.68	α	69.025	+0.008
117.68	β	69.30	-0.009
159.55	β	94.49	-0.018
159.55	γ	95.0	-0.031
159.55	α	93.99	+0.021
238.62	α	141.68	+0.001
238.62	β	142.88	+0.012
234.51	γ	141.475	-0.031
320.0	δ	194.36	+0.057
350.0	δ	213.60	-0.008

Table II. Experimental results for Pm^{151}.

H (gauss)	Transition	ν(exp) (Mc/sec)	Residuals (Mc/sec)
40.00	α	27.148	+0.002
40.00	β	28.675	-0.012
60.00	α	40.830	+0.008
60.00	β	43.120	-0.012
60.00	γ	47.135	+0.021
85.00	α	58.050	+0.037
85.00	β	61.275	-0.009
85.00	γ	66.875	-0.021
125.03	α	85.750	-0.011
125.03	β	90.575	+0.001
125.03	γ	98.790	+0.008

FIGURE LEGENDS

Fig. 1. Hyperfine-structure diagrams (schematic) for Pm^{147} and Pm^{151}.

Fig. 2. Transitions in Pm^{147}.

Fig. 3. Resonance in Pm^{147}.

Fig. 4. Transitions in Pm^{151}.

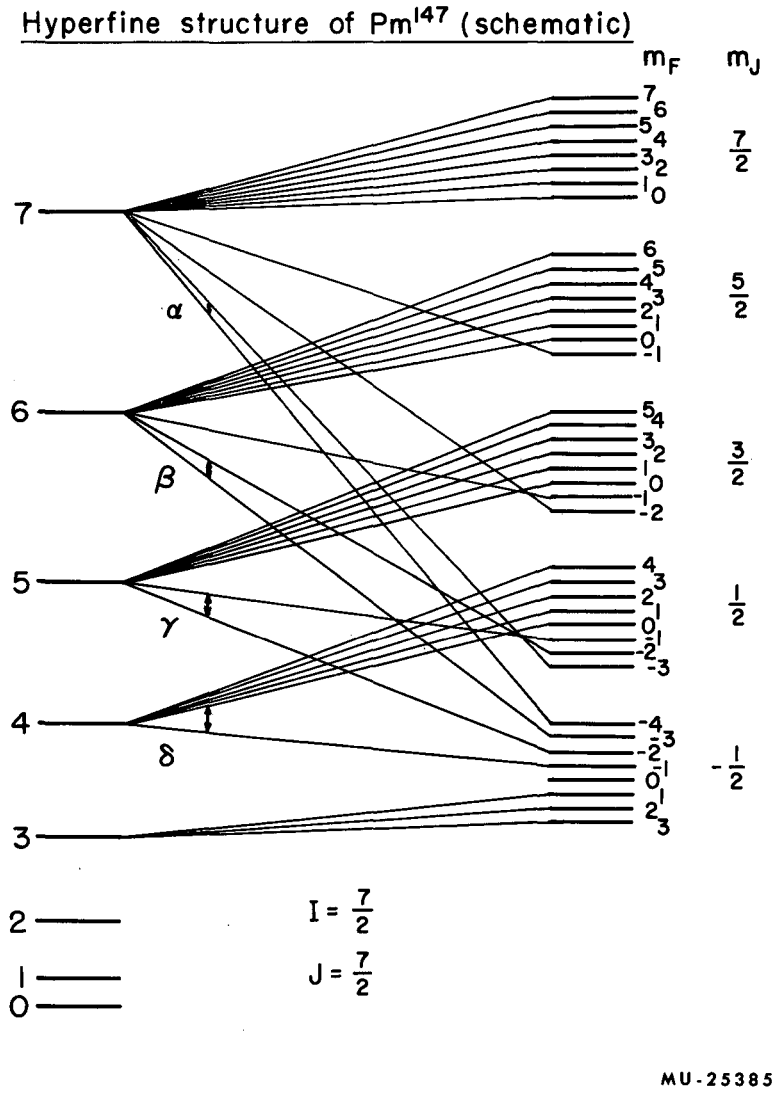

Fig. 1a. Hyperfine-structure diagrams (schematic) for Pm^{147} and Pm^{151}.

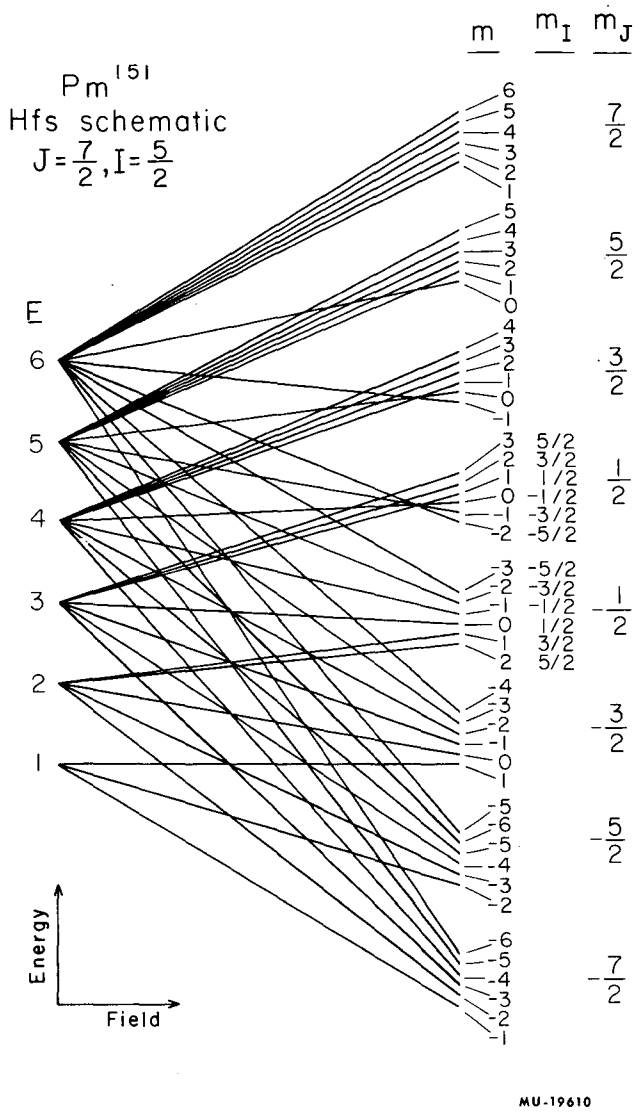
Pm151
Hfs schematic
J=7/2, I=5/2
E
6
5
4
3
2
1
m
mI
mJ
7/2
5/2
3/2
1/2
-1/2
-3/2
-5/2
Energy
Field
MU-19610

Fig. 1b

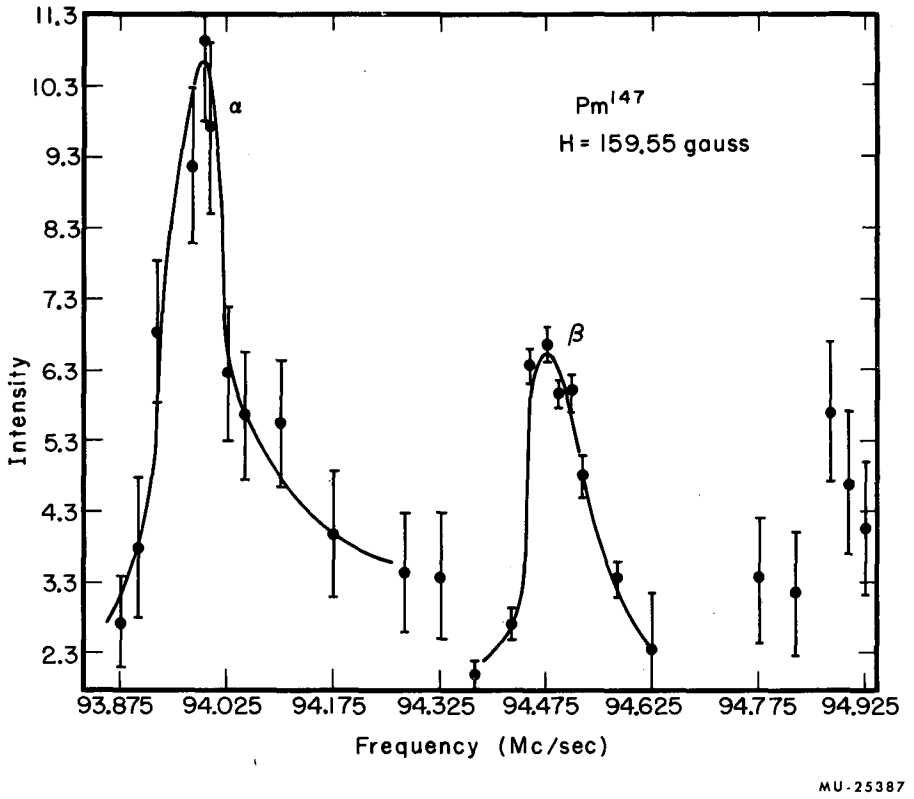

Fig. 2. Transitions in Pm^{147}.

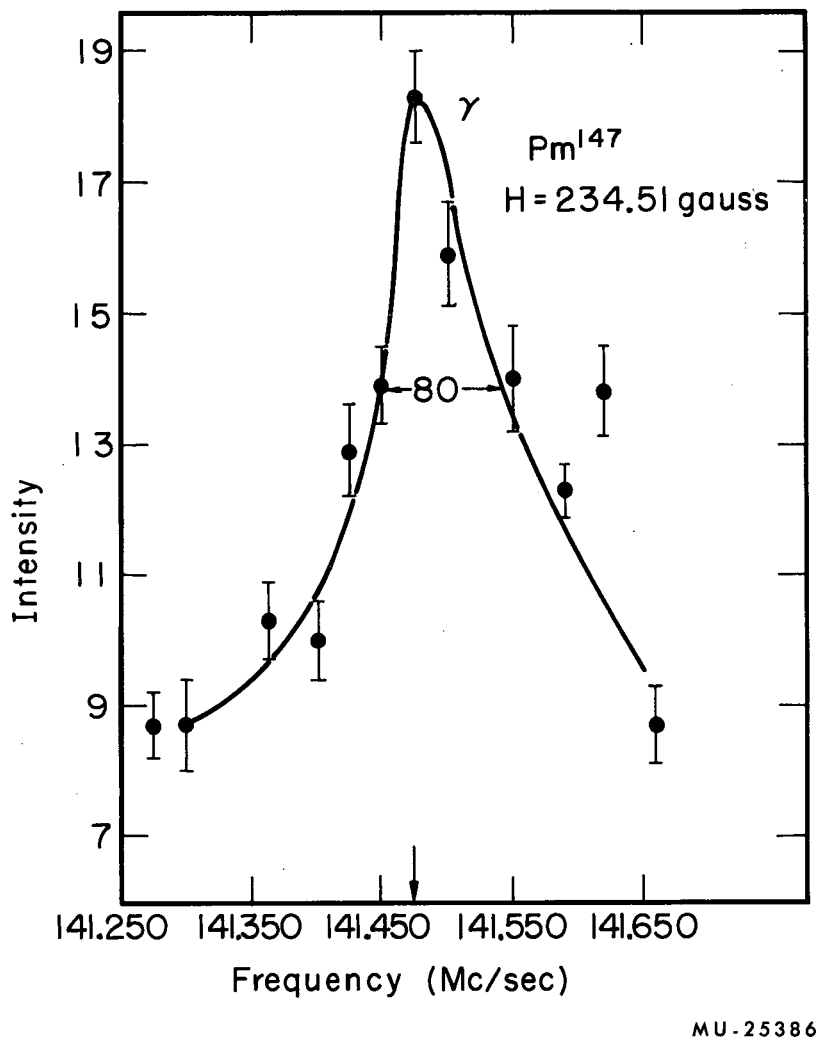

Fig. 3. Resonance in Pm^{147}.

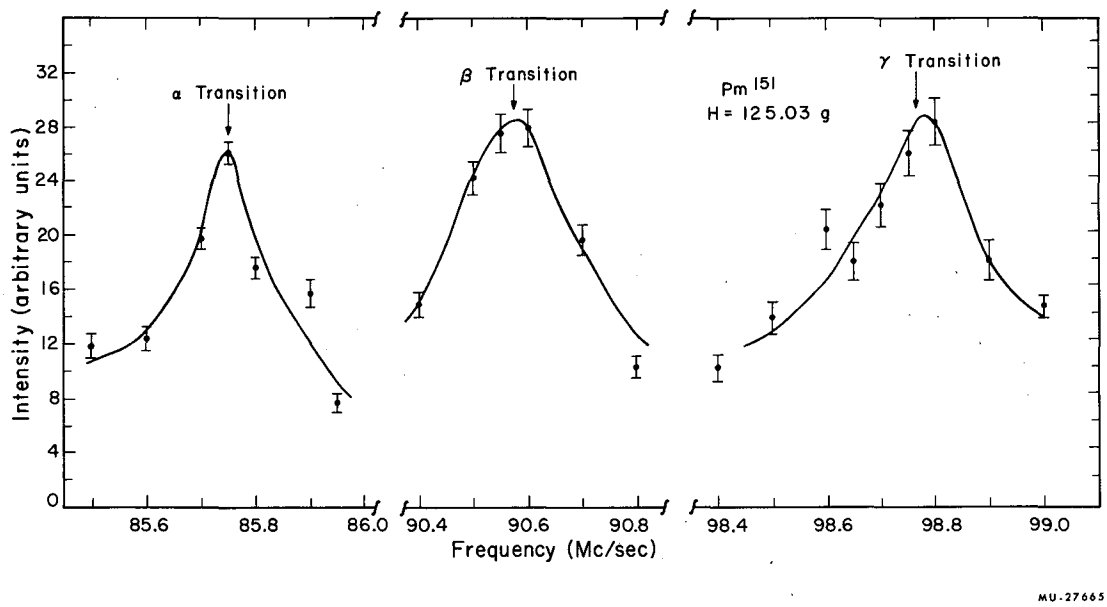

Fig. 4. Transitions in Pm^{151}.

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