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ABSTRACT

The hyperfine structure of 19-hr Pr^{142} in the electronic ground state $^4I_{9/2}$ has been studied by the atomic-beam magnetic-resonance method. The following results have been obtained: electronic splitting factor $g_J(^4I_{9/2}) = -0.7311(3)$, nuclear spin $I = 2$, magnetic-dipole hyperfine-structure constant $|A| = 67.5(5)$ Mc/sec, electric-quadrupole hyperfine-constant $|E| = 7.0(2.0)$ Mc/sec, and $B/A > 0$. From the hyperfine-structure constants, and assumptions made concerning the electronic fields at the nucleus, the nuclear moments are calculated to be $|\mu_I| = 0.297(15)$ nm, and $|Q| = 0.035(15)$ barns, with $Q/\mu_I > 0$.

HYPERFINE STRUCTURE OF PRASEODYMIUM-142^{*}

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INTRODUCTION

Precision investigations of hyperfine structure by the method of atomic beams can yield information about the electronic structure of the low-lying atomic states, and the structure of the nuclear ground state. Some features of the electronic ground state of praseodymium (Pr) have already been established by the atomic-beam work of Lew.¹ In particular, this work showed that the ground configuration of Pr is $(4f)^3$, and that coupling among the electrons to the Hund's Rule state $^4I_{9/2}$ gives good agreement with the measured electronic angular momentum (J) and electronic splitting factor (g_J). This coupling scheme seems to be characteristic of all the elements in the lanthanide series that contain 4f electrons only.² Corrections to the g_J values of systems containing 4f electrons arise from the breakdown of Russell-Saunders coupling and relativistic and diamagnetic effects. These have been recently calculated³ and have yielded, for Pr, the value $g_J = -0.7307$. As a check of this theory it seemed to us desirable to obtain a more accurate experimental value for the g_J value than that given by Lew.

Praseodymium-142 has 59 protons and 83 neutrons, and therefore lies in the region of the table of isotopes that should be well described by the shell model. On the basis of the single-particle shell model, the ground-state properties are determined by the states of the last proton

and neutron. The shell model predicts that the 59th proton should lie in the $d_{5/2}$ state. This is supported by the observed spin of Pr^{141} . The state of the 83rd neutron is very probably $f_{7/2}$, as inferred from the level-ordering scheme for neutrons and the measured spin of $7/2$ for ${}_{60}\text{Nd}^{143}$. If the proton and neutron states given above are correct assignments, then Nordheim's weak rule should apply, and the ground state spin of Pr^{142} should lie within the limits $2 \leq I \leq 6$.

A naive value of the magnetic moment (μ_I) of Pr^{142} can be obtained by assuming Schmidt values for the moments of the last odd particles, and pure J-J coupling between them. This procedure yields $\mu_I = -1.5 \text{ nm}$. Evidence concerning the moment has been obtained by Grace et al.⁴ from nuclear polarization experiments on Pr^{142} . Their experiments yield values for the moment that depend on the angular momentum (i_β) accompanying the β transition in the electron decay of Pr^{142} . They obtain

$$\mu_I = 0.11(1) \text{ nm if } i_\beta = 0$$

or

$$\mu_I = 0.17(2) \text{ nm if } i_\beta = 1,$$

which is in sharp disagreement with the value of the single-particle theory.

EXPERIMENTAL METHOD

The 19-hr isotope Pr^{142} can be produced by bombarding stable Pr^{142} with thermal neutrons. About 100 mg of pure (> 99%) praseodymium metal is encapsulated under vacuum in quartz, then irradiated for 48 hr at a flux of 9×10^{13} neutrons/cm²-sec. This produces several hundred millicuries of activity, which is sufficient for several hours of running time.

After irradiation, the sample is placed in a small, sharp-lipped tantalum crucible, which is contained in turn by a tantalum oven. The oven is placed in an oven-loader that has provision for electron-bombardment heating and can be introduced into the apparatus with minimum disturbance

to the apparatus vacuum. The apparatus is of conventional design, and employs flop-in magnet geometry according to the proposal of Zacharias.⁵ Details of the oven, oven-loader, and apparatus are given elsewhere.⁶

Detection of the radioactive praseodymium beam is accomplished by collection on freshly-flamed platinum foils. After exposure the foils are placed in methane beta counters and the deposition measured. It is estimated that the collection efficiency of platinum for praseodymium is > 50%, and very probably 100%. The result is typical of the suitability of platinum as a collector of elements throughout the lanthanide and actinide regions.

The method of taking data consists of varying the frequency at a given magnetic field. For a particular setting of the rf, a foil is exposed for 5 min. At the end of this time a new foil is placed in position and the frequency varied. The stability of the magnetic field is checked with a beam of ^{39}K that issues from an auxiliary oven in the buffer chamber. The frequency of the $\Delta F = 0$ resonance in potassium serves to calibrate the field. The intensity of the radioactive beam is calibrated between 5-min resonance exposures.

OBSERVATIONS

At low magnetic fields, the nuclear and electronic angular momenta are tightly coupled together, and the transition frequency between magnetic substates belonging to the same hyperfine level is given by

$$\nu = g_F \mu_0 H,$$

where

$$g_F \approx g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} \quad (1)$$

and a term in the nuclear moment has been neglected. Since J and g_J had been determined from the work of Lew,¹ the procedure for determining the

nuclear spin (I) consisted of exposing foils at the frequencies predicted by Eqs. (1) for possible values of I. The results of such a search are shown in Fig. 1, and clearly indicate a nuclear spin of 2.

The hyperfine structure of a system with $I = 2$ and $J = 9/2$ is indicated schematically in Fig. 2. It is seen that there are five transitions satisfying the selection rules $\Delta F = 0$, $\Delta m_F = \pm 1$ that are observable with a flop-in apparatus. Those transitions in the highest three F states (α , β , and γ , respectively) were followed up in field to about 100 gauss. Some of the observed resonances are illustrated in Fig. 3, and the data are tabulated in Table I.

At higher fields the approximation of tight coupling in Eqs. (1) is no longer valid, and the transition frequency must be determined from the Hamiltonian for the system. Within the accuracy of this experiment, the energy levels can be described by the Hamiltonian

$$\mathcal{H} = A\mathbf{I} \cdot \mathbf{J} + \frac{B[3(\mathbf{I} \cdot \mathbf{J})^2 + 3/2(\mathbf{I} \cdot \mathbf{J}) - I(I+1)J(J+1)]}{2IJ(2I-1)(2J-1)} - g_J \mu_0 (\mathbf{J} \cdot \mathbf{H}). \quad (2)$$

The first term is the magnetic-dipole interaction, the second is the electric-quadrupole interaction, and the third term gives the interaction of the electronic-dipole moment with the externally applied field (\mathbf{H}). A fit to the data is made by treating A, B, and g_J as parameters and obtaining the set of values which minimize the rms error between the observed frequencies calculated with (2). The calculation of the best fit is extremely complex. The solution for the energy of a given level involves the diagonalization of a matrix of high order. This can only be done numerically, and the task is most easily performed by means of a program for the IBM 704. This program is described elsewhere.⁷ The results of this treatment are

$$g_J = -0.7311(3) \quad |A| = 65.5(5) \text{ Mc/sec} \quad |B| = 7.0(2.0) \text{ Mc/sec.}$$

The stated errors for A and B represent standard deviations for the data. The error in g_J is chosen to be about 1 part in 2000 to allow for the possibility of systematic errors in the apparatus proportional to the magnetic field. The theoretical transition frequencies calculated with these values are compared with the experimental ones in Table I.

NUCLEAR MOMENTS

In order to infer the nuclear moments from the measured hyperfine-structure constants, the magnetic field (H_z) and the electric-quadrupole field (q_J) at the nucleus must be calculated. Such calculations can be performed using general expressions for the fields of n equivalent electrons coupled to the Hund's Rule ground term. The expression for the magnetic field has been given elsewhere,⁸ and yields

$$\langle {}^4I_{9/2} | H_z | {}^4I_{9/2} \rangle = -2\mu_0 \left\langle \frac{1}{r^3} \right\rangle_{4f} \frac{714}{121}$$

A similar expression can be derived for the quadrupole field at the nucleus. Here we state the result; the derivation is sketched in the appendix.

$$q_J = \left\langle \frac{1}{r^3} \right\rangle \langle (l)^n S = \frac{n}{2}, L = \frac{n}{2}(2l-n+1), J, m_J = J | \sum_i (3\cos^2\theta - 1) | (l)^n, S = \frac{n}{2}$$

$$L = \frac{n}{2}(2l-n+1), J, m_J = J \rangle = \mp \left\langle \frac{1}{r^3} \right\rangle \left[\frac{3K(K-1) - 4L(L+1)J(J+1)}{(2L-1)(J+1)(2J+3)} \right] \left[\frac{(2L-n^2)}{n(2l-1)(2l+3)} \right] \quad (3)$$

where $K = J(J+1) + L(L+1) - S(S+1)$.

For less than a half-filled shell, n is the number of electrons and the negative sign is used; for more than a half-filled shell, n is the number of missing electrons and the positive sign is used. Using this expression, we obtain for the quadrupole field in the ${}^4I_{9/2}$ state of Pr:

$$q_J = - \left\langle \frac{1}{r^3} \right\rangle_{4f} \frac{28}{121}$$

These results are in agreement with previous calculations of Lew.¹ We employ the value $\left\langle \frac{1}{r^3} \right\rangle_{4f} = 3.63 \text{ a}^{-3}$ that has recently been obtained by Judd and Lindgren,³ and which is estimated by them to be in error by not more than 5%. This value differs by over 25% from a value obtained by Lew,¹ from semiempirical estimates. Corrections to these values for the field will arise from the breakdown of Russell-Saunders coupling due to the spin-orbit interaction. We have calculated this effect on the fields using parameters given by Judd and Lindgren³ and find that it is less than 1%. Accordingly, we ignore this correction as small compared with the uncertainty in $\left\langle \frac{1}{r^3} \right\rangle_{4f}$.

With the stated assumptions for the fields, we obtain the nuclear moments from the measured hyperfine structure from the relations:

$$A = - \frac{1}{IJ} \mu_I \langle H_z \rangle$$

and

(4)

$$B = -e^2 q_J \Omega.$$

For the moments of Pr¹⁴² we obtain:

$$|\mu_I| = 0.297(15) \text{ nm} \quad |Q| = 0.035(15) \text{ barns} \quad \frac{Q}{\mu_I} > 0.$$

and for the moments of Pr¹⁴¹, using Lew's values¹ for the hyperfine constants,

$$|\mu_I| = 5.09(25) \text{ nm} \quad |Q| = 0.070(4) \text{ barns} \quad \frac{Q}{\mu_I} < 0.$$

DISCUSSION

From the viewpoint of the extreme single-particle model, the properties of Pr¹⁴² can be discussed in terms of the state assignments of the last proton and neutron. The state of the 59th proton has been inferred, from the measured spin of Pr¹⁴¹, to be $d_{5/2}$. The Schmidt limit for the $d_{5/2}$ state is 4.79 nm, so that with the assumptions for the magnetic field at the nucleus as stated in the previous section, the moment of Pr¹⁴¹ lies outside the Schmidt lines for spin 5/2.

Evidence for the state of the 83rd neutron can be obtained from the spins and parities of ${}_{56}\text{Ba}^{139}$ and ${}_{58}\text{Ce}^{141}$ inferred from the beta-decay studies. These measurements yield $(7/2 -)$ as the ground state of these nuclei. In addition, the spin and magnetic moment have been measured for ${}_{60}\text{Nd}^{143}$. All of these data are consistent with the assignment of $7/2$ for the 83rd neutron. We note that the measured spin of 2 is consistent with Nordheim's weak rule and the state assignments for the odd particles.

If pure J-J coupling is assumed between the proton and neutron, then the ratio of the quadrupole moments Q_{142}/Q_{141} can be predicted:

$$\frac{Q_{142}}{Q_{141}} = \frac{3C(C-1) - 4J_p(J_p+1)I(I+1)}{J_p(2J_p-1)(2I+1)(2I+3)} \quad (5)$$

where $C = I(I+1) + J_p(J_p+1) - J_n(J_n+1)$ and J_p and J_n are, respectively, the angular momenta of the proton and neutron, and I is the spin of Pr^{142} .

With the indicated state assignments, this yields:

$$\frac{Q_{142}}{Q_{141}} = -0.49,$$

which is very close to the measured absolute value of 0.50.

Similarly, we can employ the assumption of J-J coupling and values for the proton and neutron g factors inferred from the moments of Pr^{141} and Nd^{143} , respectively. This procedure yields the value -1.0 nm for the moment of Pr^{142} . This is substantially greater than the measured moment.

APPENDIX

In this section we indicate the derivation of Eq. (3) used in the text for the matrix elements of $\sum_i (3\cos^2\theta - 1)_i$ in the Hund's Rule term of maximum spin (S) and maximum orbital angular momentum (L) consistent with the Pauli principle. The state is considered to be formed by the coupling of n equivalent electrons of the configuration $(l)^n$, where n is less than a half-filled shell, i. e., $n < 2(2l + 1)$. However, the same result

holds for more than a half-filled shell, if the sign of the matrix element is changed, and if n is reinterpreted as the number of missing electrons.

We derive:

$$\left\langle \sum_1 3\cos^2\theta - 1 \right\rangle = \left\langle S = \frac{n}{2}, L = \frac{n}{2} (2l - n + 1), J, m_J = J \left| \sum_1 (\cos^2\theta - 1) \right| S = \frac{n}{2}, L = \frac{n}{2} (2l - n + 1), J, m_J = J \right\rangle. \quad (A1)$$

The Wigner-Eckart Theorem may be employed to remove the m_J dependence:

$$\left\langle \sum_1 3\cos^2\theta - 1 \right\rangle = \left[\frac{J(2J-1)}{(2J+3)(J+1)(2J+1)} \right]^{1/2} L, S, J \left\| \sum_1 (3\cos^2\theta - 1) \right\| L, S, J \rangle. \quad (A2)$$

Since $\sum_1 (3\cos^2\theta - 1)$ commutes with S , the J dependence can be removed according to the relation

$$\left\langle L, S, J \left\| \sum_1 (3\cos^2\theta - 1) \right\| L, S, J \right\rangle = \left[2J+1 \right]^{1/2} \frac{2[3K(K-1) - 4J(J+1)L(L+1)]}{[(2J-1)2J(2J+2)(2J+3)]^{1/2}} C(S, L), \quad (A3)$$

where $C(S, L)$ depends only on S and L , and $K = J(J+1) + L(L+1) - S(S+1)$.

We can determine $C(S, L)$ by combining (A2) and (A3), and evaluating $\sum_1 (3\cos^2\theta - 1)$ for the state of maximum J and maximum m_J .

Expressed in single-particle coordinates, the wave function for the state of maximum J and maximum m_J can be written as

$$\left| S = \frac{n}{2}, L = \frac{n}{2} (2l - n + 1), J = m_J = L + S \right\rangle = \left| l^+, (l-1)^+, \dots, (l-n+1)^+ \right\rangle,$$

where the notation is that of Condon and Shortley. In this way we find

$$\begin{aligned} & \left\langle l^+, (l-1)^+, \dots, (l-n+1)^+ \left| \sum_1 (3\cos^2\theta - 1) \right| l^+, (l-1)^+, \dots, (l-n+1)^+ \right\rangle \\ & = - \frac{2L(2L-n^2)}{n(2l-1)(2l+3)}, \end{aligned} \quad (A4)$$

where we have used the relation

$$\langle l, m_l | 3\cos^2\theta - 1 | l, m_l \rangle = \frac{2[3m_l^2 - l(l+1)]}{(2l-1)(2l+3)} \quad (A5)$$

Equating expression (A4) to expression (A3) leads to the result

$$C(S, L) = - \frac{(2L-n^2)}{n(2l-1)(2l+3)(2L-1)}$$

FOOTNOTES AND REFERENCES

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Table I. Summary of observations in Pr¹⁴².

Data No.	H (gauss)	Observed frequency (Mc/sec)	Obs. freq. - calc. freq. (Mc/sec)	Transition ^a
1	8.248(66)	5.837(25)	-0.018	α
2	8.248(66)	6.450(25)	+0.005	β
3	15.920(62)	11.320(30)	-0.002	α
4	15.920(62)	12.450(50)	-0.012	β
5	29.836(54)	21.300(50)	+0.006	α
6	29.836(54)	23.460(30)	+0.025	β
7	53.423(44)	38.375(50)	+0.016	α
8	53.423(44)	42.260(25)	+0.020	β
9	53.423(44)	48.412(30)	-0.052	γ
10	90.364(34)	65.475(50)	-0.063	α
11	90.364(34)	72.360(50)	+0.035	β
12	149.713(50)	110.525(50)	+0.040	α
13	149.713(50)	142.630(50)	-0.003	γ
14	279.798(29)	214.360(20)	-0.001	α
15	90.364(34)	83.240(60)	+0.004	γ

^a α : ($F = 13/2, m = -3/2$) \leftrightarrow ($F = 13/2, m = -5/2$)

β : ($F = 11/2, m = -1/2$) \leftrightarrow ($F = 11/2, m = -3/2$)

γ : ($F = 9/2, m = 1/2$) \leftrightarrow ($F = 9/2, m = -1/2$)

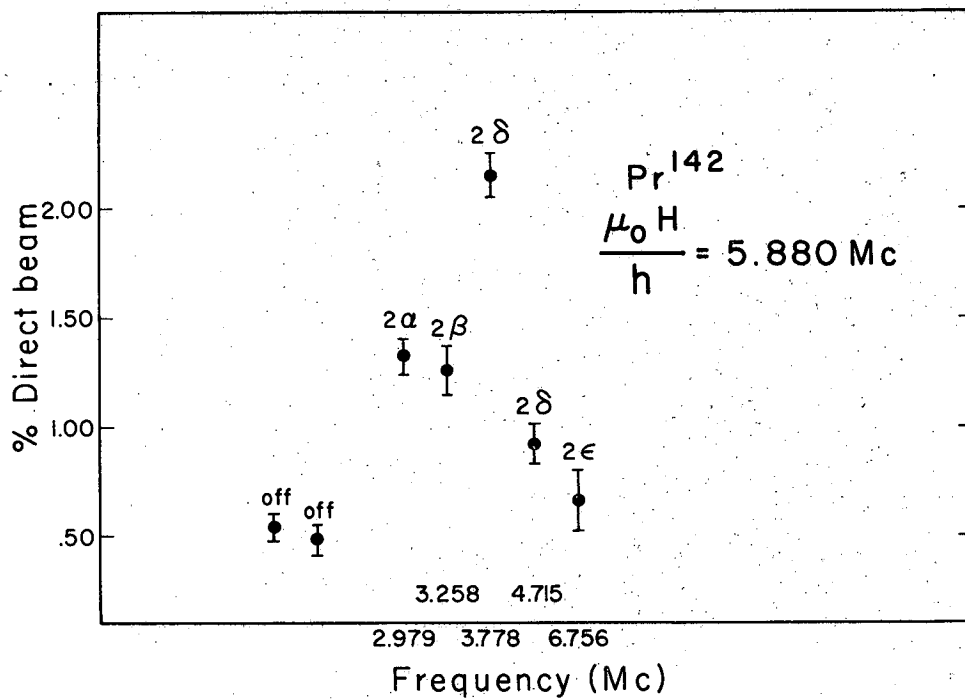
Figure Captions

Fig. 1. Results of a search at 4.2 gauss to determine the spin of Pr^{142} .

The "off" points are exposures taken with the rf turned off. The other points are exposures taken at the lower frequencies in each of the hyperfine states of $I = 2$, $J = 9/2$ with $g_J = -0.7311$.

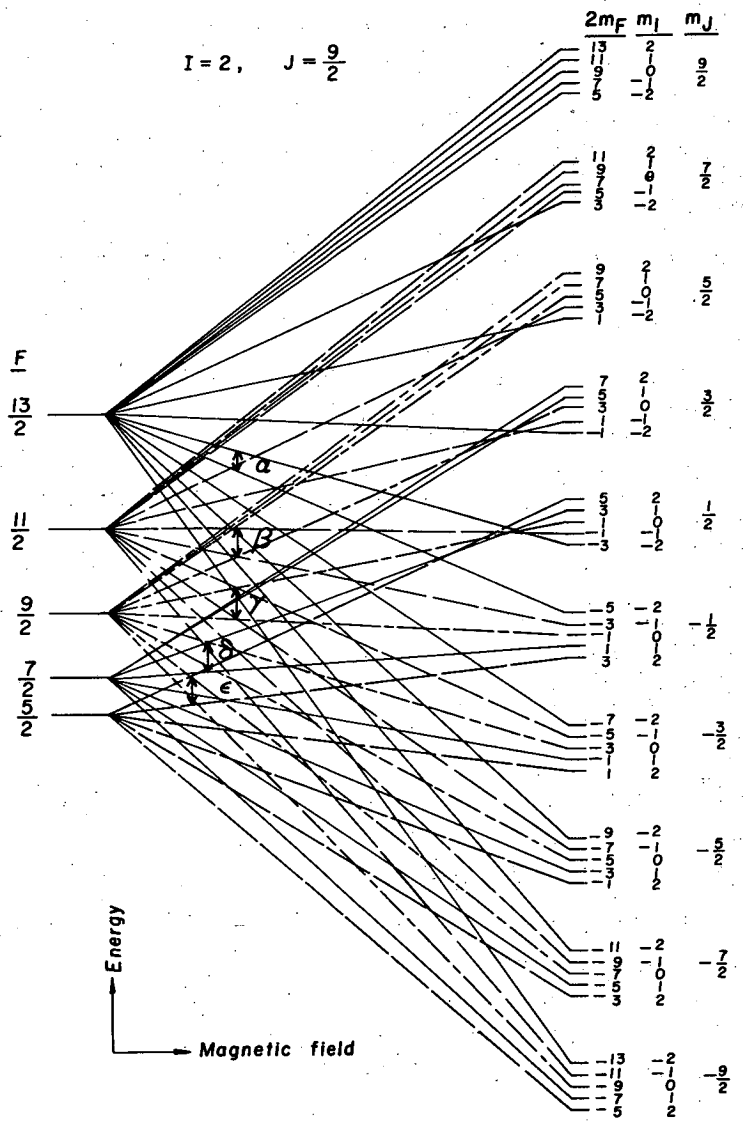
Fig. 2. Hyperfine structure of the system $I=2$, $J=9/2$ in an external magnetic field. The Greek letters denote the observable transitions in an apparatus with flop-in magnet geometry.

Fig. 3. Resonances observed in each of the three highest F states.



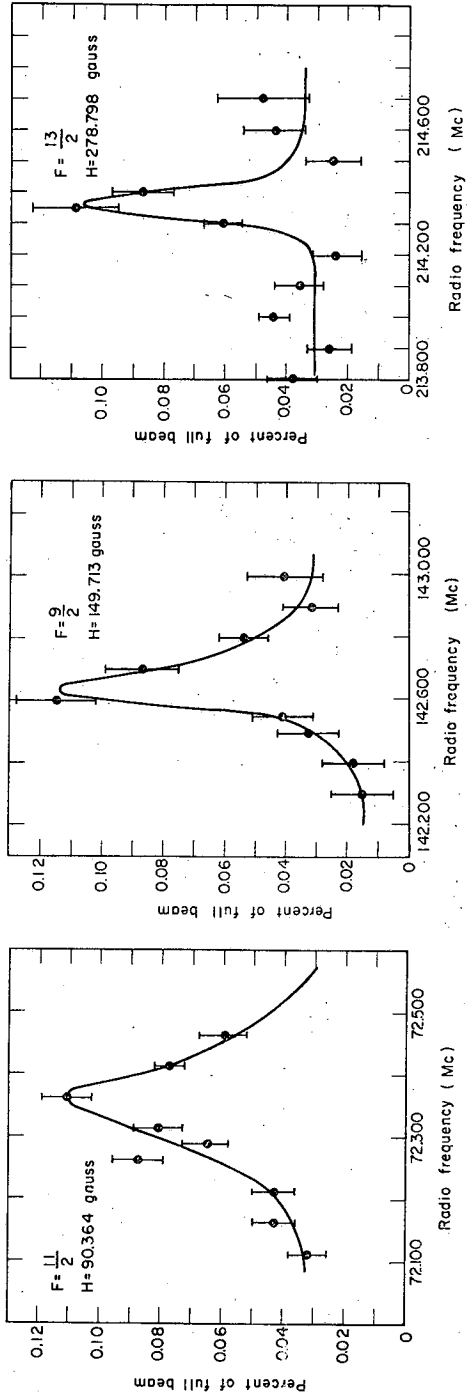
MU-17073

Fig. 1



MU-21506

Fig. 2



MUB-829

Fig. 3

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