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NUCLEAR MOMENTS OF AMERICIUM-241 AND 16-h AMERICIUM-242 AND ANALYSIS OF THE HYPERFINE FIELDS

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ABSTRACT

The nuclear moments of americium-241 and 16-h americium-242 have been directly measured by the method of triple resonance in an atomic beam. They are $\mu_I(Am^{241}) = +1.58(3)$ nm and $\mu_I(Am^{242}) = +0.3808(15)$ nm, including a correction for diamagnetic shielding. On the collective model $\mu_I(Am^{242})$ is a direct measure of the gyromagnetic ratio of the core (g_R), and the measured value of μ_I is in excellent agreement with the value $g_R = Z/A$. From these values for the magnetic moments and previous measurements of the hyperfine constants, values are deduced for the magnetic fields at the nucleus. It is shown that breakdown of Russell-Saunders coupling including relativistic effects gives contributions of the wrong sign to the magnetic field. Arguments are given which show that the residual effect is most probably due to core polarization.

INTRODUCTION

In this paper we describe direct measurements of the magnetic moments of americium-241 and americium-242 by the method of triple resonance in an atomic beam. These measurements are of interest for two reasons. First, the nuclear ground state of americium-242 has been shown to be characterized by $K = 0$.[1] Therefore, the magnetic moment of americium-242 is a direct measure of the rotational gyromagnetic ratio of the core (g_R). Second, the electronic ground state of Am is a half-filled 5f shell, $(5f)^7$, for which the Hund's rule state is $^8S_{7/2}$. To this approximation the hyperfine fields should vanish. Hence the values of the hyperfine fields are measures of the sizes of relativistic effects, of core polarization effects, and of the amount of configuration mixing present in the ground state wave function.

Previous atomic beam work on americium-241 (Ref. 1) and americium-242 (Ref. 2) had established the hyperfine constants of the ground state. In particular, the electronic angular momentum (J), the splitting factor (g_s), the magnetic dipole hyperfine constant (A), and the electric quadrupole constant (B) were determined. Hence, a direct measurement of the nuclear moment coupled with the measured A value can give a direct measure of the electronic field at the nucleus (H_z) through the relation $A = -\frac{1}{IJ}\mu_I H_z$.

EXPERIMENTAL METHODS AND RESULTS

Americium-241 in an HCl solution was obtained from the stockpile of the Lawrence Radiation Laboratory group headed by Professor Burris Cunningham. Americium oxide was made from the solution by adding NH_4OH and heating the precipitate $[Am(OH)_3]$ in a furnace until oxidation occurred.

The atomic-beam oven used was of Ta, with a Ta inner liner. The americium oxide was placed in the oven with an excess of lanthanum metal. When the oven was heated to approximately 1000°C, the lanthanum reduced the Am_2O_3 to Am metal. The reduction proceeded very slowly, however, requiring several hours.

The experimental method used was identical to that reported previously on rhenium.[3] Signal-to-noise ratios of 3:1 were obtainable with the hairpins singly and also with all three hairpins together. The hyperfine levels of $Am^{241}(I=5/2)$ and $Am^{242}(I=1)$ are shown schematically in Figs. 1 and 2, respectively. The A and B hairpins were set on the transitions labeled a, the C hairpin on the transitions labeled by Arabic numerals. Sample resonances are shown in Fig. 3.

The data obtained for Am^{242}, which consist of one high-field single-hairpin transition and six triple-loop transitions, were combined with those of Ref. 1 for the purpose of data reduction. A least-squares fit to the data was obtained with a Hamiltonian of the form

$$A(\underset{\sim}{I}\cdot\underset{\sim}{J}) + B\frac{3(\underset{\sim}{I}\cdot\underset{\sim}{J})^2 + 3/2(\underset{\sim}{I}\cdot\underset{\sim}{J}) - I(I+1)J(J+1)}{2IJ(2I-1)(2J-1)} - g_J\mu_0\underset{\sim}{J}\cdot\underset{\sim}{H} - g_I\mu_0\underset{\sim}{I}\cdot\underset{\sim}{H}\,, \qquad (1)$$

with A, B, g_J, and g_I as variable parameters.

Difficulty was encountered in observing high-field a transitions in Am^{241} at the frequencies predicted by using the results of Ref. 1. A low-field direct transition was observed, confirming the value of A and B. Using these values of A and B, and g_J of Am^{242} to predict resonance frequencies, we were able to observe high-field α transitions. The data obtained in this experiment, one direct transition and six triple-loop transitions, were combined with the previous direct transition data for data reduction. These data were also fitted to a Hamiltonian of the form of Eq. (1), but this time only A, B, and g_I were varied, with g_J fixed at the value of g_J found in Am^{242}.

Final results were:

In Am^{241},

$$A = \pm\, 17.1437(0.0028)\ \text{Mc/sec},$$

$$B = \mp\, 123.8477(0.0323)\ \text{Mc/sec},$$

$$g_I = 3.42(0.06) \times 10^{-4}.$$

In Am^{242},

$$A = \pm\, 10.1282(0.0014)\ \text{Mc/sec},$$

$$B = \pm\, 69.6339(0.0013)\ \text{Mc/sec},$$

$$g_J = -\,1.937884(0.000067),$$

$$g_I = 2.059(0.008).$$

This leads to a hyperfine anomaly of

$$^{241}\Delta^{242} = 1.7(2.0)\%\ .$$

Correcting the values of g_I for diamagnetic shielding, using the expression $g_I = g_I^{\text{screened}}/(1-\sigma)$ and the value of σ corresponding[4] to $Z = 65$, gives

$$g_I(241) = 3.45(0.06) \times 10^{-4},$$

$$\mu_I(241) = 1.58(0.03)\ \text{nm},$$

$$g_I(242) = 2.074(0.008) \times 10^{-4},$$

$$\mu_I(242) = 0.3808(0.0015)\ \text{nm}.$$

Because both measured μ_I's have the same sign, both values of A must also have the same sign.

An investigation was made to see if second-order electronic perturbation could contribute significantly to A or g_I. It was found that such effects are negligible.

HYPERFINE FIELDS

In a nonrelativistic treatment, A and B are zero in an atom having a half-filled closed shell coupling to a Hund's rule state. Marrus, Nierenberg, and Winocur[1] investigated the breakdown of L-S coupling in Am in an effort to explain the measured values of A and B. Their analysis yielded the ground state wave function

$$\Psi(J=7/2) = 0.882\,|{}^8S_{7/2}\rangle + 0.457\,|{}^6P_{7/2}\rangle - 0.114\,|{}^6D_{7/2}\rangle \ .$$

Nonrelativistic values of A and B obtained by Marrus et al.,[1] using this wave function, are given in Table I. The magnitude of the results is in agreement with the experimental results, but the sign of the ratio A/B is in error. We have considered two other sources which could contribute to A and B: (a) relativistic effects and (b) core polarization.

Bordarier, Judd, and Klapisch[5] showed that A for a configuration ℓ^N of Dirac electrons is given by

$$A = \frac{1}{I}\,\frac{\langle J\|X\|J\rangle}{\langle J\|J\|J\rangle} \ ,$$

where

$$X = a_{10}W^{(10)1} + a_{01}W^{(01)1} + a_{12}W^{(12)1} \ .$$

The a_{ij}'s depend only on the electronic relativistic radial wave functions and the ℓ of the electrons. The double tensor $W^{(\kappa k)K}$ is defined as a sum over the n electrons

$$W^{(\kappa k)K} = \sum_{i=1}^{n} w_i^{(\kappa k)K} ,$$

$$w^{(\kappa k)K} = (t^{\kappa} v^{k})^{K} ,$$

where

$$\langle s \| t^{\kappa} \| s \rangle = [\kappa]^{1/2} ,$$
$$\langle \ell \| v^{k} \| \ell' \rangle = [k]^{1/2} \delta_{\ell \ell'} .$$

The double tensors above can be simply related to familiar operators

$$W^{(01)1} = \left[\frac{3}{(2\ell)(\ell+1)(2\ell+1)}\right]^{1/2} \underset{\sim}{L} ,$$
$$W^{(10)1} = \left[\frac{2}{2\ell+1}\right]^{1/2} \underset{\sim}{S} ,$$
$$W^{(12)1} = - \sum_{i=1}^{n} \left[\frac{10(2\ell-1)(2\ell+3)}{(\ell)(\ell+1)(2\ell+1)}\right]^{1/2} (sC^2)_i^1 .$$

Writing $S = L + 2S - J$, $L = 2J - (L + 2S)$, and $N_i = \ell_i - (10)^{1/2} (sC^2)_i^1$, we obtain

$$A(\mathrm{rel}) = (-g_J - 2)\alpha + \beta + \gamma \frac{\langle J \| \sum_i N_i \| J \rangle}{\langle J \| J \| J \rangle} ,$$

where

$$\alpha = \frac{2\mu_N e}{I(2\ell+1)^2} \left[\frac{2}{3}(2\ell-1)(\ell+1) P_{++} - \frac{8}{3}\ell(\ell+1) P_{+-} + (\frac{4}{3}\ell^2 + 2\ell) P_{--}\right] ,$$

$$\beta = -\frac{8}{3} \frac{\mu_N e}{I(2\ell+1)^2} \left[(\ell+1)^2 P_{++} + \ell(\ell+1) P_{+-} + \ell^2 P_{--}\right] ,$$

$$\gamma = \frac{2}{3} \frac{\mu_N e}{I(2\ell+1)^2} \left[4(\ell+1)(2\ell-1) P_{++} - (2\ell-1)(2\ell+3) P_{+-} + 4\ell(2\ell+3) P_{--}\right] ,$$

$$P_{++} = \int \frac{F_+ G_+}{r^2}\, dr ,$$

$$P_{+-} = \int \frac{F_+ G_- + G_+ F_-}{r^2}\, dr ,$$

$$P_{--} = \int \frac{F_- G_-}{r^2}\, dr .$$

F_+, G_+ are radial wave functions for $j = \ell + 1/2$; F_-, G_-, for $j = \ell - 1/2$. N_i is the angular operator appearing in the nonrelativistic expression for A, and $\gamma \dfrac{\langle J \| \sum_i N_i \| J \rangle}{\langle J \| J \| J \rangle}$ is nearly one times the nonrelativistic A value.

Bordarier et al.[5] also found that B for the configuration ℓ^n of Dirac electrons is given by

$$B = 2e^2 Q \begin{pmatrix} J & 2 & J \\ -J & 0 & J \end{pmatrix} \langle J \| Z^2 \| J \rangle .$$

Z^2 is given by

$$Z^2 = b_{11} W^{(11)2} + b_{13} W^{(13)2} + b_{02} W^{(02)2} ,$$

with

$$b_{11} = - \left[\frac{4\ell(\ell+1)}{25(2\ell+1)^3} \right]^{1/2} \left[-(\ell+2) R_{++} + 3R_{+-} + (\ell-1) R_{--} \right],$$

$$b_{13} = - \left[\frac{6(\ell-1)(\ell)(\ell+1)(\ell+2)}{25(2\ell-1)(2\ell+1)^3(2\ell+3)} \right]^{1/2} \left[(2\ell+1) R_{++} + 4R_{+-} - (2\ell+3) R_{--} \right],$$

$$b_{02} = \left[\frac{2\ell(\ell+1)}{5(2\ell-1)(2\ell+1)^3(2\ell+3)} \right]^{1/2} \left[(2\ell-1) R_{++} + 6R_{+-} + (\ell-1)(2\ell+3) R_{--} \right],$$

$$R_{++} = \int \frac{F_+^2 + G_+^2}{r^3}\, dr ,$$

$$R_{+-} = \int \frac{F_+F_- + G_+G_-}{r^3} \, dr ,$$

$$R_{--} = \int \frac{F_-^2 + G_-^2}{r^3} \, dr .$$

The radial integrals were evaluated by use of the relativistic Am wave functions of Liberman, Waber, and Cromer.[6] These integrals are tabulated in Table II.

The wave functions of Liberman et al. are very effective in predicting atomic energy levels.[7] It is well known, however, that wave functions which predict energy eigenvalues well are often useless for predicting hyperfine structures. For this reason it is wise to try to estimate the validity of these wave functions when they are used to evaluate the integrals above. One parameter closely related to these integrals is ζ, the spin orbit coupling constant, since all are proportional to $\langle 1/r^3 \rangle$. The difference in energy eigenvalues for $f_{7/2}$ and $f_{5/2}$ electrons should be $7/2\,\zeta$; these wave functions then give $\zeta(\mathrm{Am}) = 3020\ \mathrm{cm}^{-1}$. Blume, Freeman, and Watson[8] showed that, in the rare earths, ζ obtained from a Hartree-Fock calculation is decreased by about 10% if two-body interactions such as spin-other-orbit are considered. If such a factor holds for Am, then the value of ζ given by these wave functions would be lowered to approximately $2700\ \mathrm{cm}^{-1}$; the correct value of $\zeta(\mathrm{Am})$ is approximately $2400\ \mathrm{cm}^{-1}$. A ζ calculated from these wave functions would then be of the order of 12% too large. Pryce and Foglio, from an investigation based on the Thomas-Fermi model, found that in the region of Pu and Am, $\zeta / \langle 1/r^3 \rangle \approx 370\ \mathrm{cm}^{-1}/a_0^{-3}$. When the correct value of ζ given above is used, this relation gives

$\langle 1/r^3 \rangle = 6.5\, a_0^{-3}$. We can also calculate $\langle 1/r^3 \rangle$ from $1/r^3 = \int \frac{F^2 + G^2}{r^3}\, dr$, obtaining $\langle 1/r^3 \rangle \approx 8.1\, a_0^{-3}$. This value is about 20% higher than that obtained from the relationship suggested by Pryce and Foglio.

Unfortunately there is no way of estimating whether the ratios of the various radial integrals are in error. It does appear that the magnitudes of the integrals might be too high by about 15%. If such is the case, the relativistic values we obtain below for A and B should be lowered by 15%, with corresponding changes in the amount of core polarization.

In Table I we give the relativistic corrections to the nonrelativistic A and B values. These corrections are obtained by use of the full radial integrals given in Table II. The calculated and measured values of B agree fairly well if we assume the measured value of B is positive. The A values, on the other hand, are in very poor agreement. If we assume that core polarization is responsible for the discrepancy between the calculated and measured values, we obtain

$$\Delta A = -28.2 \frac{\mu_I}{I}, \qquad -81.6 \frac{\mu_I}{I} \quad \text{Mc/sec},$$

where the first number above holds if the measured A's are positive, the second if the A's are negative. These values would be reduced to $-20.0\, \mu_I/I$, $-73.4\, \mu_I/I$ if the calculated A values were decreased by 15%.

A value of ΔA can also be obtained from core polarization values in Pu. Bauche and Judd[9] showed that

$$\frac{\Delta A(\text{Am})}{\Delta A(\text{Pu})} \approx \frac{[g_J(\text{Am}) + 1]\, \mu_I(\text{Am})\, I(\text{Pu})}{(g_J(\text{Pu}) + 1)\, \mu_I(\text{Pu})\, I(\text{Am})} .$$

Armstrong,[10] on the basis of an analysis of the A values of the first six excited states of Pu, concluded that $\Delta A(\text{Pu}) \approx -12$ Mc/sec. This leads to

$\Delta A(Am) \approx -61 \frac{\mu_I}{I}$ Mc/sec, which agrees reasonably well with the values obtained above for A less than zero, and in particular with the value obtained by using the decreased A value.

It is conceivable that some mechanism, such as quadrupole shielding, exists which would change the sign of the calculated value of B. We feel, however, that this is unlikely and that the sign of $B(Am^{241})$ is positive and $B(Am^{242})$ is negative (see below). This forces the sign of A for both isotopes to be negative, which agrees with the sign of A obtained from the Pu core polarization.

NUCLEAR STRUCTURE

The spin of Am^{241} is known[11] to be $I = 5/2$. After investigating the α decay of Am^{241} to Np^{237}, Stephens, Asaro, and Perlman[12] concluded that the unpaired 95th proton must be in the Nilsson orbital 5/2-[523]. This assignment fits the Nilsson energy level diagram[13] exactly if $0.21 < \delta < 0.28$. One can also obtain a value for the deformation from the optically measured quadrupole moment $Q(241) = +4.9$ barns;[11] the derived value of $\delta = 0.21$ supports the proposed proton orbital assignment.

The magnetic moment μ_I of Am^{241} has been calculated by using the Nilsson wave functions. Table III shows the results of this calculation, which was performed for several positive values of δ by using both free nucleon g factors and quenched g factors.[14] The value $g_R = Z/A$ was used. We see that, if we use free nucleon g factors, the measured moment is predicted at $\delta \approx 0.15$; if we use quenched g factors, at δ slightly greater than 0.2. The result obtained by using quenched g factors is consistent with that previously obtained.

In Am^{242}, the odd neutron is probably in the Nilsson orbit

5/2+[622]. This assignment, which corresponds to $0.22 < \delta < 0.26$, is also made for the odd neutron in the ground states of the isotones Pu^{241} and Cm^{243}. Using the coupling rules of Gallagher and Moszkowski,[15] we then have $K = \Omega_P - \Omega_N = 0$ for Am^{242}. For a $K = 0$ nucleus, $\mu_I = g_R I$; if we accept the proposed value of K, we then have a direct measurement of the core g factor. The measured value of g_R, or μ_I, is 0.381, to be compared to the usually used value of $g_R = Z/A = 0.392$.

Because $K = 0$ in Am^{242}, we have

$$Q(242) = -\frac{1}{5}Q_0 ,$$

where Q_0 is the intrinsic quadrupole moment. If δ is positive as indicated by the level assignment, Q_0 is positive. Q(242) will then be negative, causing B(241) and B(242) to have different signs.

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FOOTNOTES AND REFERENCES

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Table I. Contributions to the hyperfine constants A and B in americium.

	Magnitude A (Mc/sec)	Magnitude B (Mc/sec)
Breakdown of LS coupling within $(5f)^7(7s)^2$	$26.4\,\frac{\mu_I}{I}$[a]	$27.9\,Q$[b]
Relativistic corrections	$28.5\,\frac{\mu_I}{I}$	$0.3\,Q$
Core polarization	$-81.6\,\frac{\mu_I}{I}$	0
Total calculated	$-26.7\,\frac{\mu_I}{I}$	$28.2\,Q$
Total measured	$-26.7\,\frac{\mu_I}{I}$	$25.3\,Q$

[a] μ_I in nm.

[b] Q in barns.

Table II. Am relativistic radial integrals (in units of a_0^{-3}).

$e\int \frac{F_+G_+}{r^2}\,dr$	=	$-23.5\ \mu_0$
$e\int \frac{F_-G_-}{r^2}\,dr$	=	$31.6\ \mu_0$
$e\int \frac{F_+G_- + F_-G_+}{r^2}\,dr$	=	$6.7\ \mu_0$
$\int \frac{F_+^2 + G_+^2}{r^3}\,dr$	=	7.6
$\int \frac{F_-^2 + G_-^2}{r^3}\,dr$	=	8.6
$\int \frac{F_+F_- + G_+G_-}{r^3}\,dr$	=	8.2

Table III. Am^{241} nuclear moments calculated with Nilsson wave functions.

	η		
	2	4	6
Free nucleon g factors	1.89	1.32	1.07
Quenched g factors	1.95	1.58	1.41

Proton state 5/2 - [523].

FIGURE CAPTIONS

Fig. 1. Breit-Rabi diagram for Am^{241}.

Fig. 2. Breit-Rabi diagram for Am^{242}.

Fig. 3. Some observed triple resonances in Am.

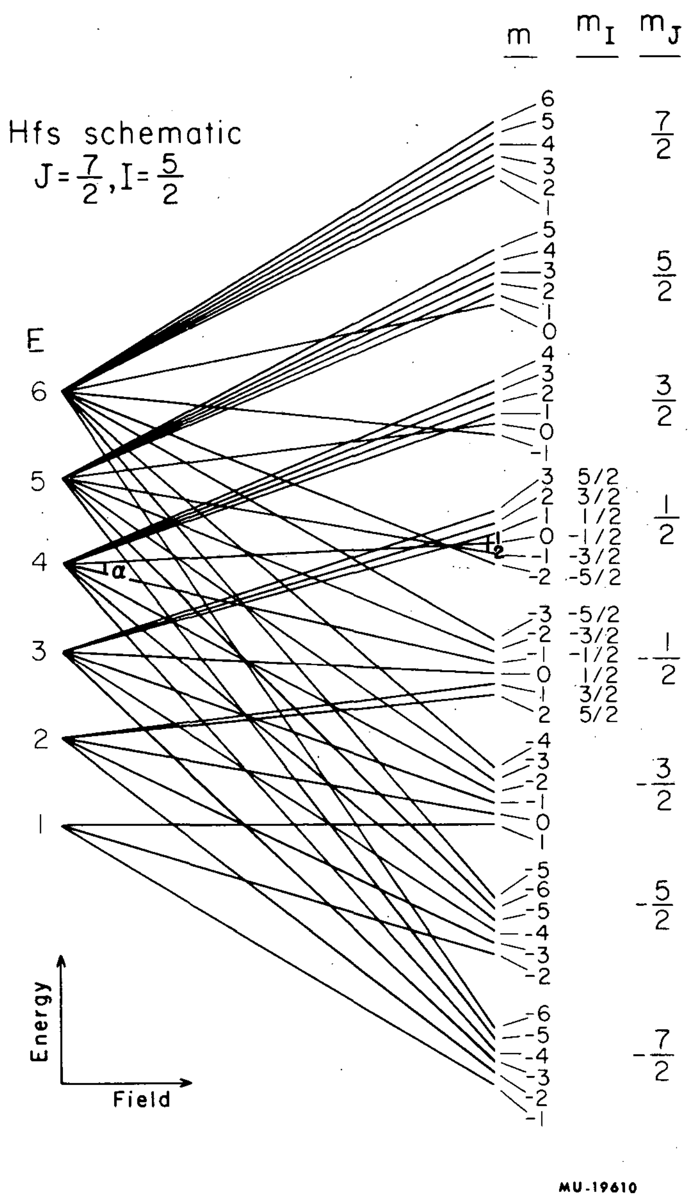

Fig. 1

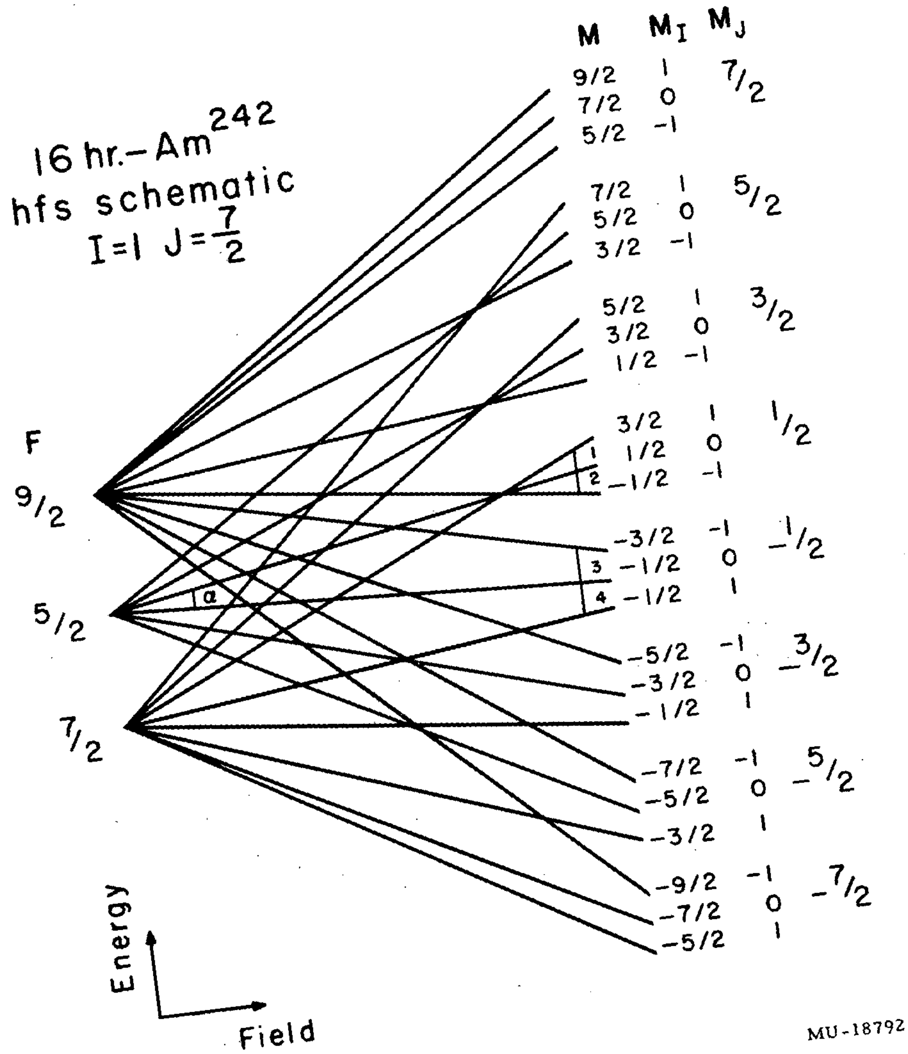
16 hr.–Am242
hfs schematic
I=1 J=7/2
M
MI
MJ
F
9/2
5/2
7/2
Energy
Field
MU-18792

Fig. 2

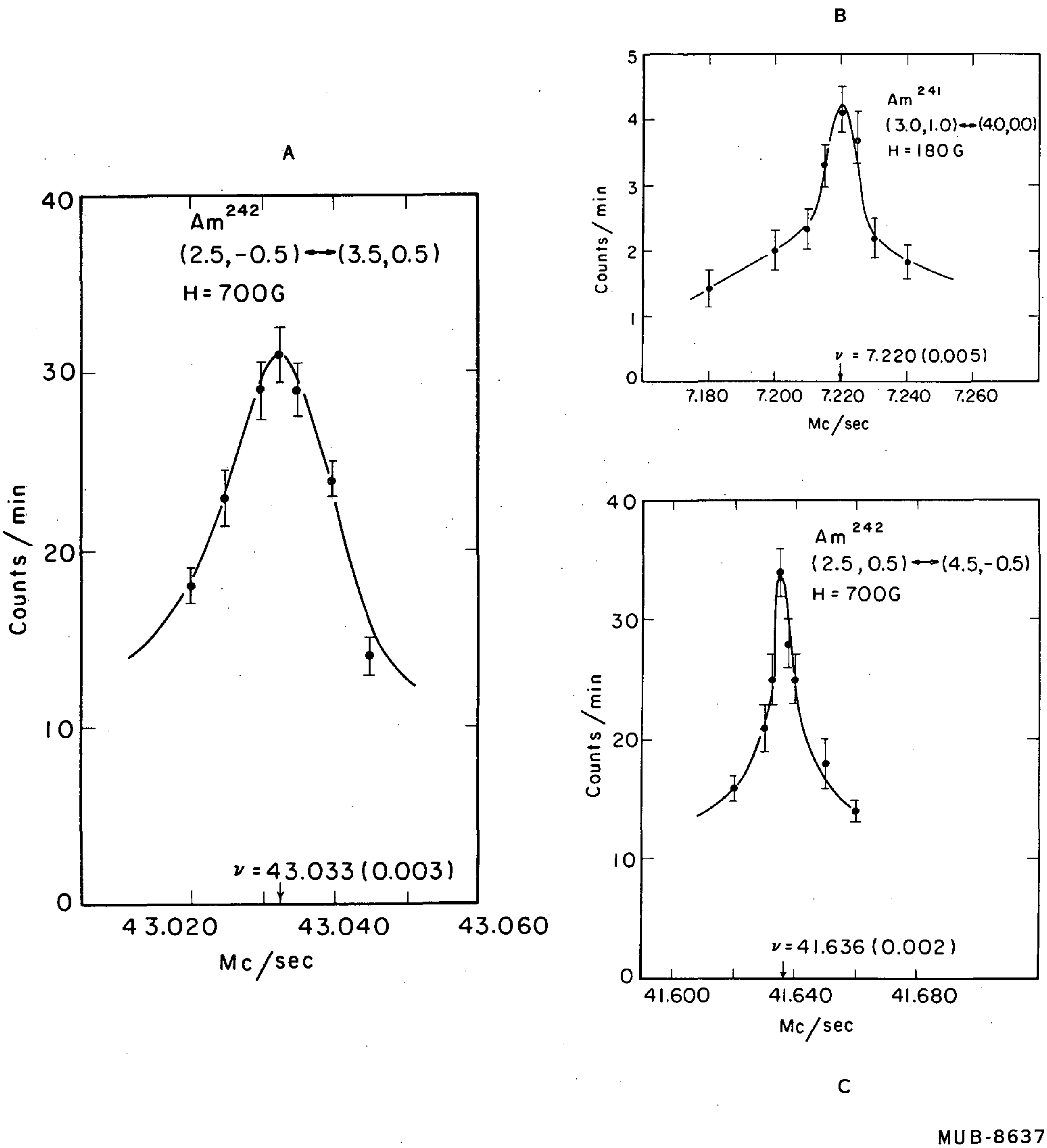
A
Am^242
(2.5,-0.5)⟷(3.5,0.5)
H=700G
ν=43.033 (0.003)
Counts / min
Mc/sec
B
Am^241
(3.0,1.0)⟷(4.0,0.0)
H=180G
ν=7.220 (0.005)
Counts / min
Mc/sec
C
Am^242
(2.5,0.5)⟷(4.5,-0.5)
H=700G
ν=41.636 (0.002)
Counts / min
Mc/sec
MUB-8637

Fig. 3