New Mössbauer Levels in the Rare Earths Following Coulomb Excitation*

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By using Coulomb excitation to populate the excited states of the nuclei, new Mössbauer levels have been observed in Gd^{160}, Dy^{164}, and Er^{168}. Mössbauer levels have been observed following Coulomb excitation in Gd^{155}, Gd^{156}, and Gd^{158}. For the even-even isotopes of gadolinium, hyperfine structures are poorly resolved, but under appropriate assumptions relative quadrupole moments can be obtained which agree with the values derived from Coulomb excitation cross-section data. Well-resolved magnetic hyperfine structure was observed in Er^{168}, and the magnetic energy splitting of $(7.8\pm0.4)\times10^{-6}$ eV was obtained. In the case of Dy^{164} a well-resolved hyperfine structure was observed, but the interpretation of the complex structure was not definite.

I. INTRODUCTION

THE application of Mössbauer spectroscopy in the past has been limited to a restricted class of nuclei. Previously, in order to carry out a Mössbauer experiment, it was necessary that there exist a suitable radioactive parent that could populate the excited states of the nucleus being studied. This parent nucleus had to be sufficiently long-lived to permit the Mössbauer measurements to be carried out. The state under investigation had to be sufficiently strongly populated to give an acceptable signal-to-noise ratio. These requirements have limited the number of potential Mössbauer nuclei for which Mössbauer studies were possible, and have precluded the use of Mössbauer spectroscopy for carrying out systematic studies of nuclear properties. It would, for example, be of interest to determine directly the variation of such quantities as the nuclear-quadrupole moment or gyromagnetic ratio in a series of isotopes of a given element without recourse to nuclear models. Considerations of this type have motivated the search for other means than β decay of populating excited states for carrying out Mössbauer studies.

In the past year, the feasibility of employing nuclear reactions or Coulomb excitation for populating the excited states of nuclei and subsequently observing recoilless emission of the γ rays produced in the deexcitation of these states has been well established.[1–11] Of the various techniques, Coulomb excitation has proved to be the most useful. Detailed studies[12] following Coulomb excitation of Fe^{57} in a metallic lattice have shown that the observed Mössbauer fraction and magnetic hyperfine interaction are the same as when the excited states of Fe^{57} are populated by electron capture from Co^{57}. Further studies with Fe_2O_3 have shown that in the oxide lattice a reduction in the Mössbauer fraction is observed following Coulomb excitation, but that the same magnetic hyperfine interaction is present as when a Co^{57} source is used to populate the excited states of Fe^{57}. Thus the use of Coulomb excitation would appear to provide a general method for populating the excited states in a series of isotopes and subsequently carrying out Mössbauer measurements.

There are two regions of the periodic table where such studies are expected to be particularly useful, namely, in the rare earths and in the actinides. In these regions there are many low-lying nuclear levels suitable for Mössbauer experiments. Moreover, in the rare-earth region the electronic configurations have an incomplete $4f$ shell which gives rise to large internal fields at the nucleus in many of the rare-earth compounds. Thus, one would expect to see well-resolved hyperfine structures in Mössbauer absorption spectra if the nuclei being studied can be put in suitable environments.

In the rare-earth region the ground-state deformations become very large and low-lying collective states having large Coulomb-excitation cross sections are observed well below any intrinsic levels. For example, in Fig. 1 one can see that there are five isotopes of gadolinium where there are suitable levels for recoilless emission. Of these isotopes only the levels in Gd^{155} could

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readily be studied[13,14] using a β-decaying parent. If a more general method for populating the excited states for Mössbauer experiments were possible, all these isotopes could be studied.

In the present work Coulomb excitation is used to populate excited states, and subsequently Mössbauer studies are carried out in various isotopes of three rare-earth elements. A detailed study was made of the gadolinium isotopes, and an effort is made to study any trend in the systematic variation of the nuclear-quadrupole moment of three of the even-even isotopes. Well-resolved magnetic hyperfine structure was observed in Dy^{164} and Er^{168}, and estimates of magnetic energy splitting were obtained. Indications of distortions in the absorption spectra of Er^{168} and Dy^{164} have been observed which may be attributed to radiation-damage effects in target.

II. EXPERIMENTAL CONSIDERATIONS

A. Target Cryogenic System

The low-lying nuclear levels in the rare-earth region have energies which are typically 50 to 100 keV. In order to obtain sufficiently large Mössbauer fractions to study detailed hyperfine structures, both target and absorber must be cooled to liquid-helium temperature. In the present application the cryogenic system must permit the introduction of charged-particle beams onto the cooled target. The necessity of cooling the target and absorber in such experiments gives rise to several cryogenic problems. The target must be able to withstand continual bombardment by charged-particle beams for several days without disintegrating. Further, it must be maintained at a temperature close to liquid-helium temperature while beam powers of the order of a watt are to be dissipated. The Mössbauer absorber must also be cooled to liquid-helium temperature, and some means of introducing a specified relative velocity between the target and absorber must be employed. A schematic diagram showing the general layout of the cryogenic target-chamber system and the details of the helium cold finger and Mössbauer drive assembly are presented in Figs. 2 and 3, respectively. Details of this system will be published later.

B. Dectector and Electronics

The Mössbauer spectra were obtained with an integral line-scintillation detector which consisted of a NaI(Tl) scintillation crystal $\frac{1}{4}$ in. thick by 2.5 in. diam. coupled to an 8054 photomultiplier tube via a quartz light pipe. This detector was inserted into a cylindrical housing (see Fig. 2) which protruded into the cubical target chamber in such a way that the scintillation crystal was only 1 in. from the Mössbauer absorber. Standard modular electronics were used for the pulse counting and logic functions. A 256-channel pulse-height analyzer operated in the time mode with a multiscaling logic unit was used to collect and store the Mössbauer absorption spectra.

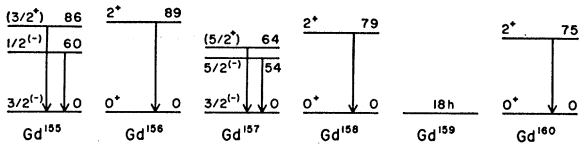

FIG. 1. Low-lying levels in the gadolinium isotopes.

C. Mössbauer Absorbers and Drive Mechanism

The Mössbauer absorbers used in these experiments were fabricated from either metallic foils or powders of suitable compounds of the rare earths considered. Materials of natural isotopic abundance were used. The materials were bonded to a copper mounting plate with a commercial epoxy resin, and thin copper foils were placed over the absorber to protect it from charged particles scattered from the target. The thickness of the copper mounting plate was chosen to obtain optimal attenuation of the lower-energy K x-rays, which were the principal source of background in these experiments. Plate thicknesses from 15 to 40 mils were employed, depending on the element and the energy level being studied. The absorber mounting plate assembly (see Fig. 3) was attached to the helium cold finger by means of thin beryllium-copper flexure reeds, and thermally grounded to it by means of copper grounding braid.

The absorber was nominally $\frac{7}{8}$ in. from the target and was capable of vibration amplitudes of $\pm\frac{1}{4}$ in. This amplitude permitted Mössbauer absorption spectra to be taken with a maximum velocity range of 3.60 cm/sec at a drive frequency of 3.8 cps (at which frequency most of these experiments were carried out). Higher velocity ranges were employed in some experiments by using a higher driving frequency.

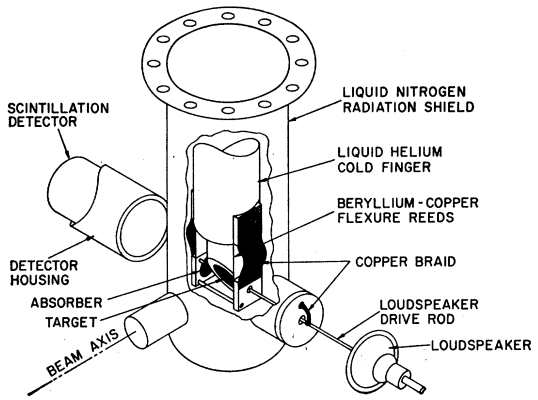

FIG. 2. Schematic diagram of the cryogenic target chamber employed in Mössbauer experiments following Coulomb excitation.

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[14] C. L. Herzenberg, L. Meyer-Schutzmeister, L. L. Lee, Jr., and S. S. Hanna, Bull. Am. Phys. Soc. **7**, 39 (1962).

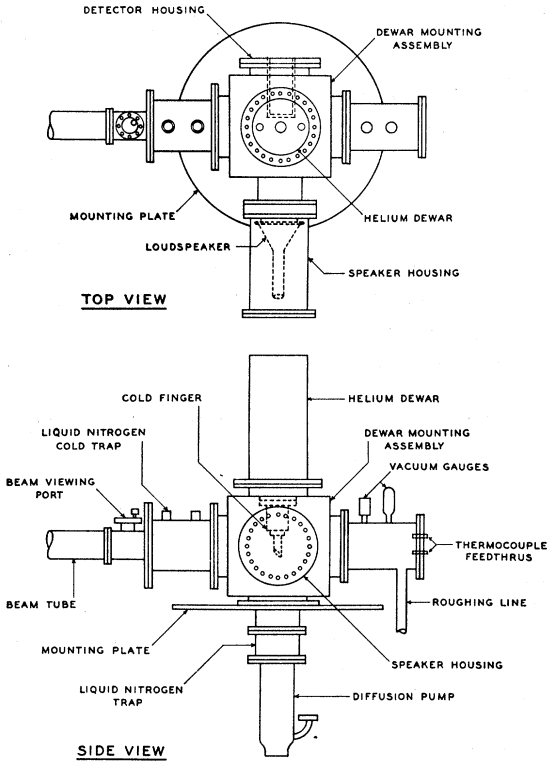

FIG. 3. Schematic diagram of the details of the helium cold finger and Mössbauer absorber assembly.

The absorber was vibrated with a parabolic displacement by coupling it with a long stainless-steel drive rod to a conventional loudspeaker driver unit located approximately 15 in. away. The loudspeaker and drive circuit used to produce the parabolic displacement were similar to those described by Cohen.[15] The loudspeaker driver was enclosed in the vacuum system in order to shield it from vibration in the Van de Graaff area and to preclude problems in feeding mechanical motion into the vacuum system. Since the loudspeaker and helium Dewar were rigidly mounted onto the same mechanical structure, the system was relatively insensitive to external vibrations. No difficulty has been observed in operating the loudspeaker in a hard vacuum (10^{-6} Torr). This system has been used to obtain Mössbauer spectra over a range of maximum velocities from 2 mm/sec to 10 cm/sec.

The drive system was calibrated at a number of velocities by observing the magnetic hyperfine splitting in Fe^{57}. The velocity calibrations obtained have an accuracy better than 1%.

D. Targets and Charged-Particle Beam

The targets used in these experiments were enriched sesquioxides of the rare-earth isotopes studied. These oxide targets were employed because of their high Debye temperatures and their ready availability. The oxide powder was bonded to the cryogenic target holder by means of thin films of epoxy resin. These targets darkened appreciably during the runs, presumably due to the cracking of carbon as a result of the bombardment, but no decrease in counting rate was observed, and no significant difference was seen in the data taken at different times during a run.

The possibility of using metallic foils for targets was considered, but at the time of the experiments such enriched foils were not available, and the detector used did not permit resolution of the various lines which would be produced if natural foils were used.

The total γ-ray yield for a thick target is approximately a factor of 5 greater for proton bombardment than for α-particle bombardment in the present case. Since it was desirable to optimize total γ-ray yield for a given beam current and thus a given rate of liquid-helium consumption, proton beams were used with relatively thick (30 mg/cm^2) oxide targets. The observed counting rates for the rare earths studied were typically of the order of 1000 counts/sec in the γ-ray window, and typical runs required several days of continuous running time on the Van de Graaff.

III. HYPERFINE INTERACTIONS

In the sesquioxides the rare-earth ions occupy nonequivalent lattice sites, with three-quarters of the ions occupying sites of C_2 symmetry and the other quarter occupying sites of C_{3i} symmetry. It is sufficient to consider only the C_2 sites[16] in intepreting the quadrupole interactions; the presence of ions in the C_{3i} sites appears to lead to only a broadening of the absorption lines. Since the internal fields are difficult to determine accurately, it is useful in studying nuclear systematics to carry out a series of experiments with different isotopes in the same crystalline lattice; then the ratios of moments can be obtained from appropriate considerations of the observed energy splittings. In order to obtain absolute values of these moments, the relevant internal fields must be determined independently, either by calculation or by additional experimental evidence.

The nuclear hyperfine interaction results from the interaction of the electric-quadrupole moment Q with the electric field gradient V_{zz}, and the magnetic dipole moment $\boldsymbol{\mu}$ with the magnetic field $\mathbf{H}$ at the nucleus. In the considerations that have been made in the present study with the even-even rare-earth isotopes, it has been assumed that the magnetic field lies along the principal axis of the electric-field-gradient tensor having the largest component, and this common direction (z axis) is chosen as the axis of quantization. The interaction Hamiltonian, giving the magnetic interaction $\mathcal{H}_M$ and the electric-quadrupole interaction $\mathcal{H}_Q$, is then

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given by

$$\mathcal{H}=\mathcal{H}_M+\mathcal{H}_Q=-\boldsymbol{\mu}\cdot\mathbf{H}+\frac{eQV_{zz}}{4I(2I-1)}\times[3I_z^2-I(I+1)+\eta(I_x^2-I_y^2)],$$

where $\eta=(V_{xx}-V_{yy})/V_{zz}$ is the asymmetry parameter and I is the nuclear spin operator. The eigenvalues of this Hamiltonian give the nuclear hyperfine splitting. For the even-even nuclei, the first excited state has spin 2, so that this level will split into five levels, and since the ground state is spin 0, all transition probabilities from these excited state levels are equal.

For the case of an axially symmetric electric field gradient, the energy eigenvalues are given by

$$E=g\mu_nHm+[eQV_{zz}/4I(2I-1)][3I_z^2-I(I+1)],$$

where g is the gyromagnetic ratio, μ_n is the nuclear Bohr magnetron, and m is the magnetic quantum number.

For pure quadrupole interaction and for $I=2$, the energy eigenvalues are given by

$$E_1(0,2,-2)=\tfrac{1}{4}eQV_{zz}(1+\tfrac{1}{3}\eta^2)^{1/2},$$
$$E_2(0,2,-2)=-\tfrac{1}{4}eQV_{zz}(1+\tfrac{1}{3}\eta^2)^{1/2},$$
$$E_3(2,-2)=\tfrac{1}{4}eQV_{zz},$$
$$E_4(1,-1)=\tfrac{1}{4}eQV_{zz}(-\tfrac{1}{2}+\tfrac{1}{2}\eta),$$
$$E_5(1,-1)=\tfrac{1}{4}eQV_{zz}(-\tfrac{1}{2}-\tfrac{1}{2}\eta),$$

where the arguments denote the eigenvalues of the eigenstates of I_z which mix to form the corresponding eigenvectors.

The magnetic energy splitting is given by $g\mu_nH$, and the characteristic quadrupole-interaction energy for $I=2$ is conventionally taken as $\frac{1}{4}eQV_{zz}$.

IV. EXPERIMENTAL RESULTS

A. Data Analysis

Mössbauer absorption spectra which exhibit various types of hyperfine structures have been observed following Coulomb excitation in Gd^{155}, Gd^{156}, Gd^{158}, Gd^{160}, Dy^{164}, and Er^{168}. Additional Mössbauer spectra have also been obtained for Gd^{155} using a $Eu^{155}{}_2O_3$ radioactive source with oxide, metallic, and intermetallic gadolinium absorbers. In all these experiments the data analysis has been carried out using a least-squares fitting routine, and the interaction energies involved have been obtained.

In those cases where well-resolved hyperfine structure was observed, computer fits were made assuming independent Lorentzian absorption lines. In those cases where hyperfine structure was not well resolved, efforts are nevertheless made to unfold the spectra under best available information concerning the nature of hyperfine structure of the particular nucleus. The results of computer fits are such that the present analysis gives highly convincing sets of numbers for hyperfine parameters for gadolinium data. In the computer analysis a constrained set of absorption lines has been assumed in which the relative magnitudes and positions of the absorption lines were fixed in accordance with the types of interaction assumed to be present, while the linewidths, depths, and interaction energies were varied in order to obtain the best fit. Several models of the interactions assumed to be present were considered. The values of quadrupole-interaction energy obtained were relatively insensitive to the forms of the other interactions assumed to be present and did not differ by more than 3% in the fits that have been made.

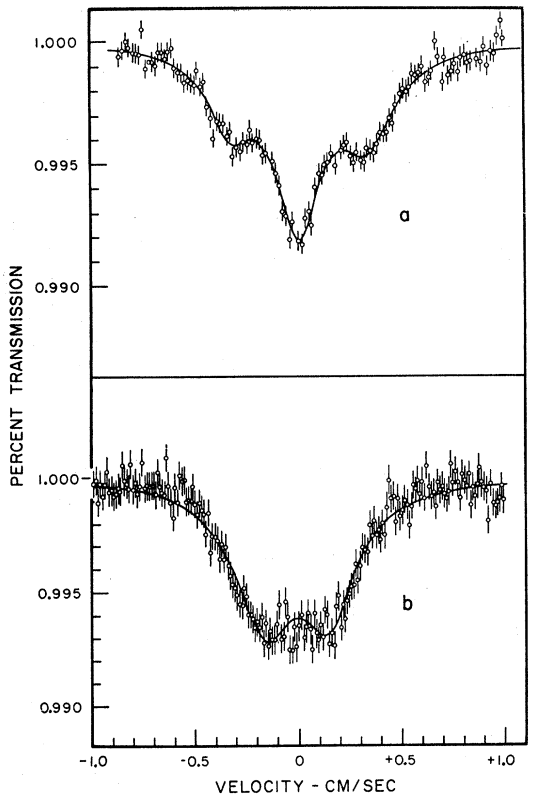

FIG. 4. Mössbauer absorption spectra for the 86.5-keV state of Gd^{155} at 4.2°K for (a) Gd_2O_3 absorber with 10.7 mg/cm² of Gd^{155}; (b) metallic Gd absorber with 31 mg/cm² of Gd^{155}. These data have been corrected for nonresonant background.

B. Gd^{155} Results Using a $Eu^{155}{}_2O_3$ Radioactive Source

Recoilless emission and absorption of the γ radiation from the second excited state of Gd^{155} at 86.5 keV was observed following β decay of Eu^{155}, and partially resolved hyperfine structure was seen in the absorption spectrum. The preliminary results have already been published.[13] The Mössbauer effect has been observed previously[14] in the 86.5-keV state without resolution of structure in the absorption spectrum, and interesting results which were obtained at liquid-nitrogen tempera-

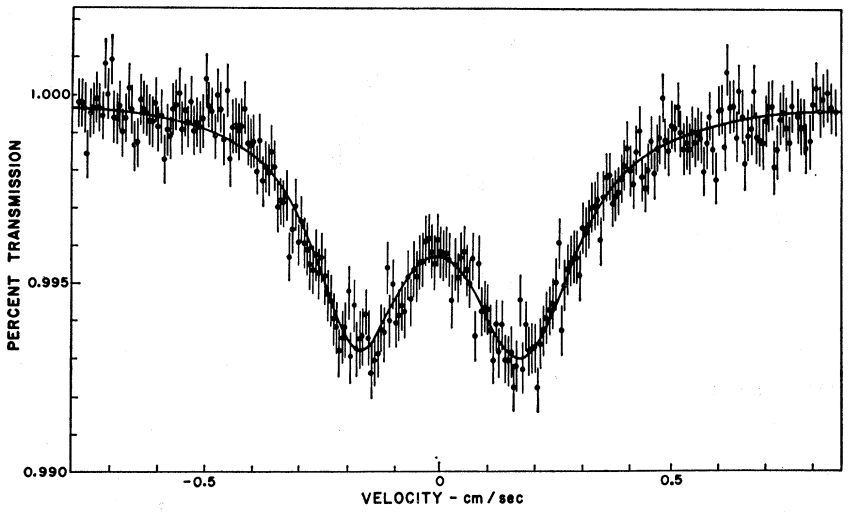

FIG. 5. Mössbauer absorption spectra for the 86.5-keV state of Gd^{155} at 4.2°K for a $GdAl_2$ absorber containing 15 mg/cm² of Gd^{155}. These data have been corrected for nonresonant background.

ture were recently published.[17] The present observations have been carried out with oxide, metallic, and intermetallic absorbers, and the energy splittings observed for the quadrupole coupling of the ground state have been used with the interaction energies obtained from the spectra of the even-even isotopes to obtain relative quadrupole moments.

The Mössbauer absorption spectra observed for the 86.5-keV state using oxide and metallic absorbers exhibit partially resolved hyperfine structure (see Fig. 4). A three-line fit was made to the oxide-absorber data, which gave 1:2:1 intensity ratios with equal splitting (3.3 mm/sec) for the two side absorption peaks. The result is indicative of two source lines overlapping two absorber lines with the same splitting. A two-line fit was made to the metallic-absorber data, which gave 1:1 intensity ratios with a splitting of 3.1 mm/sec. A Mössbauer absorption spectrum was also observed for this state using a $GdAl_2$ absorber which exhibited well-resolved splitting of the two lines seen with the oxide and metallic absorbers (Fig. 5). A two-line fit was made to this spectrum, giving intensity ratios of 1:1 with a splitting of 3.6 mm/sec. The results of these experiments to calibrate the absorbers are presented in Table I, together with the quadrupole coupling energies extracted from the observed energy splittings. Since the ground-state spin of Gd^{155} is $\frac{3}{2}$, the ground-state quadrupole coupling energy is

$$\Delta E_Q{}^{155}=\tfrac{1}{2}eQV_{zz}(1+\tfrac{1}{3}\eta^2)^{1/2}.$$

TABLE I. Mössbauer absorber calibrations using a $Eu^{155}{}_2O_3$ source.

Absorber	mg/cm² of Gd^{155} Absorber thickness	$E_Q{}^{155}$ (eV)[a] ($\times 10^{-6}$)
Gd_2O_3	10.7	(0.95±0.02)
Metallic Gd	31.0	(0.90±0.02)
$GdAl_2$	18.0	(1.03±0.01)

[a] Uncertainty is statistical on the basis of least-squares fit to assumed hyperfine interaction forms. The mean deviation, $\pm 0.07 \times 10^{-6}$ eV, of the three determinations is a more realistic uncertainty.

The errors listed in Table I are the statistical errors in fitting the spectra. Since rather simplified models have been used in analyzing these spectra, neglecting the two sites in the oxide and the magnetic hyperfine structure, the accuracy of the results obtained cannot be expected to be as good as the experimental precision of the measurements. Perhaps the best assessment of the accuracy of the results is given by the variation of the splitting energy among different absorbers which is as large as ±7%.

The ground state of Gd^{155} is known to be $\frac{3}{2}$.[18] For the 86.5-keV state a Nilsson model assignment of $\frac{3}{2}$ has been made on the basis of the 5-nsec half-life.[19] There is also indirect evidence that this state has a $\frac{5}{2}$ spin.[17,20,21] The present result of a two-line Mössbauer spectrum obtained with a $GdAl_2$ absorber can be accounted for by the sizeable ground-state quadrupole splitting alone without assuming any splitting for the 86.5-keV state of a magnitude comparable to the ground-state splitting. The present result that the 86.5-keV level shows little structure prompted us to say in our previous communication[13] that the spin assignment to this level is more likely to be $\frac{1}{2}$. However, the present result can be reconciled with the assignment of $\frac{5}{2}$ if one assumes that the quadrupole moment of this state is a small fraction of the quadrupole moment of the ground state. It is rather unlikely that the 86.5-keV state with a spin of $\frac{3}{2}$ would have a negligible structure in the face of the fact that

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the ground state with the same spin of $\frac{3}{2}$ has a clear doublet structure.

C. Gd^{155} Results Following Coulomb Excitation

Mössbauer studies have also been carried out in Gd^{155} following Coulomb excitation, and a Mössbauer absorption spectrum has been observed for the 60-keV state at both liquid-helium and liquid-nitrogen temperatures (see Fig. 6). An enriched sesquioxide target of Gd^{155} was bombarded with a 0.15-μA beam of 3-MeV protons. A natural Gd_2O_3 absorber containing 10.7 mg/cm^2 of Gd^{155} was used. The counting rate obtained in the γ-ray window was 650 counts/sec. The 60-keV γ-ray peak was barely observable in the wings of the more intense Gd K x rays. An attempt was made to extract this peak from the background, and an estimate of the signal-to-noise ratio of 0.2 ± 0.1 was obtained. A single absorption dip of 1.5% was observed at liquid-helium temperature and a similar absorption dip of 0.8% was observed at liquid-nitrogen temperature.

In previous work in this laboratory[13] the Mössbauer effect has been seen for the 60-keV state using a $Eu^{155}{}_2O_3$ source; a linewidth of 2.9 cm/sec was observed, whereas the linewidth obtained with the same absorber in the same geometry following Coulomb excitation of Gd^{155} was 3.8 cm/sec. This broadening can be accounted for entirely by resonant self-absorption in the target, which was approximately three radiation lengths thick.

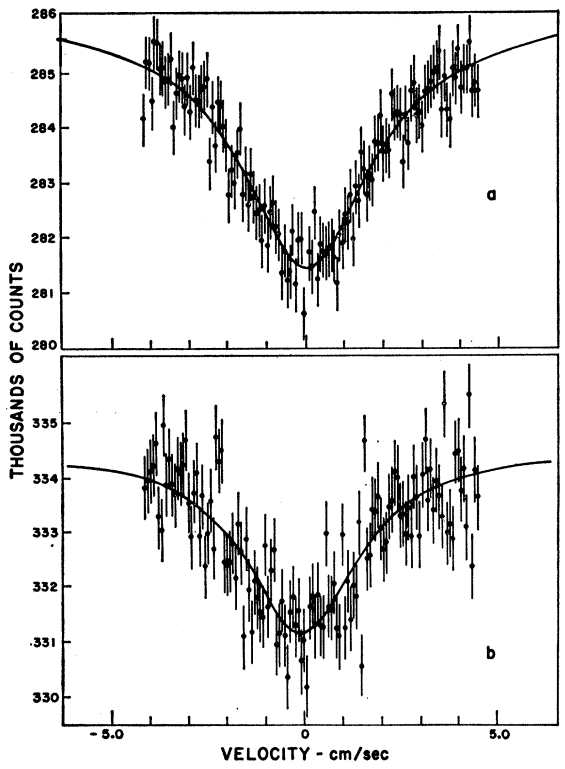

FIG. 6. Mössbauer absorption spectra following Coulomb excitation of the 60-keV state of Gd^{155} at (a) 4.2°K and (b) 77°K. A Gd_2O_3 absorber was used which contained 10.7 mg/cm^2 of Gd^{155} No background corrections have been made to these data.

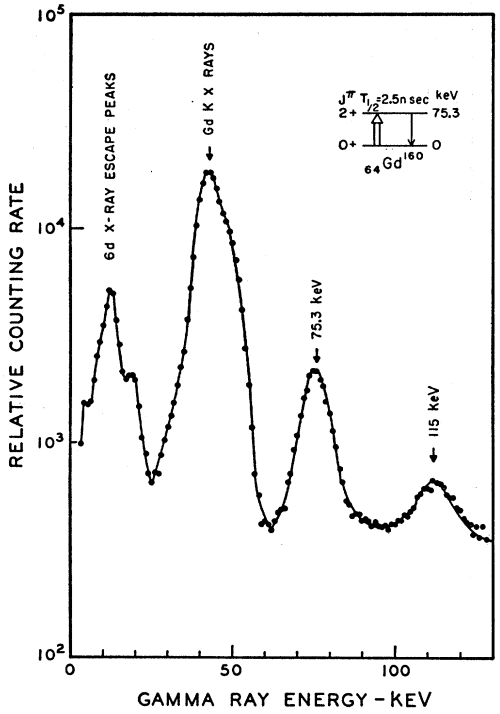

FIG. 7. Gamma-ray spectrum following Coulomb excitation of $Gd_2{}^{160}O_3$.

An estimate of the Mössbauer fraction was obtained for this state from the Coulomb-excitation data using the method of Shirley *et al.*[22] It was assumed that the spectrum observed could be considered as a single absorption line with a natural linewidth given by that observed with the radioactive source. The effect of resonant self-absorption in the target was taken into account by evaluating the transmission integrals given by Margulies *et al.*[23] for this case. It was found that the experimental Mössbauer fraction was 0.6 of the intrinsic Mössbauer fraction in this experiment and that, after making this correction, the observed intrinsic Mössbauer fraction was 0.23 ± 0.10. More accurate determinations of this f value would be possible with better resolution of the γ-ray line.

D. Results in the Even-Even Isotopes of Gadolinium

Recoilless emission and absorption were observed following Coulomb excitation of the first excited states in Gd^{156}, Gd^{158}, and Gd^{160}. Enriched sesquioxide targets of approximately 30 mg/cm^2 were bombarded with 0.2-μA beams of 3-MeV protons. The Coulomb-excited γ rays were well separated from the gadolinium K x rays, and counting rates of the order of 1000 counts/sec were

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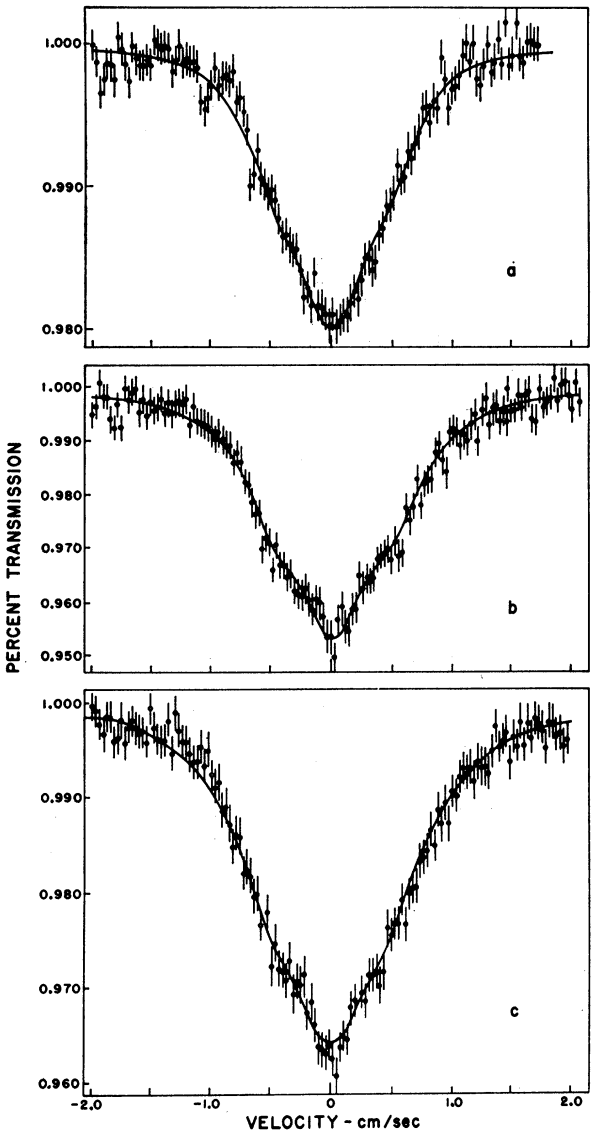

FIG. 8. Mössbauer absorption spectra following Coulomb excitation for (a) 89.0-keV state of Gd^{156} with 15 mg/cm² of Gd^{156} absorber; (b) 79.5-keV state of Gd^{158} with 18 mg/cm² of Gd^{158} absorber; and (c) 75.3-keV state of Gd^{160} with 16 mg/cm² of Gd^{160} absorber. These spectra were taken at 4.2°K with the same Gd_2O_3 absorber. These data have been corrected for nonresonant background.

achieved, with a signal-to-noise ratio of 4:1. A typical Coulomb-excitation γ-ray spectrum is presented in Fig. 7. The Mössbauer absorption spectra observed are shown in Fig. 8. The broadening in the wings of these absorption spectra is attributed to unresolved quadrupole splitting in the first excited states of these isotopes. Computer fits were made to extract the quadrupole-interaction energies.[24] The results of these computer fits are presented in Table II.

[24] In order to effect a comparison of these data with the Gd^{155} data, the quadrupole-interaction energies which have actually been extracted for these isotopes are $E_Q{}^A=\frac{1}{4}eQV_{zz}(1+\frac{1}{3}\eta^2)^{1/2}$. Since the value of η generally found in the rare-earth sesquioxides is of the order of 0.3, this quadrupole-interaction energy is not significantly different from that of the conventional definition, $\frac{1}{4}eQV_{zz}$.

In previous work Fink and Kienle[7] have also observed the Mössbauer effect in Gd^{156} and Gd^{158} following neutron capture in Gd^{155} and Gd^{157}, respectively. Their results are similar to the results obtained in the present work, except that the magnitudes of the effects they observed were somewhat less than those observed in the present work.

Additional Mössbauer spectra were also obtained for Gd^{160} using metallic and intermetallic absorbers in order to obtain better resolution of the quadrupole structure in Gd^{160}. The spectra obtained are presented in Fig. 9, and the quadrupole-interaction energies extracted are presented in Table II. The data taken with the $GdAl_2$ absorber exhibit two closely spaced absorption lines which are interpreted as partially resolved quadrupole structure in the oxide target. Attempts to fit these data, including magnetic interaction on the basis of the approximate model assumed, were not successful; the computer fit shown in Fig. 9 assumed that the absorber was unsplit and that the target exhibited pure quadrupole splitting.

The errors listed in Table II are the statistical errors in fitting the spectra. Here again the same remarks made in connection with Table I apply. Neglecting the asymmetry of the electric field gradient, the effect of two sites in oxide, and the magnetic hyperfine structure makes the uncertainty represented by the statistical errors unrealistic. However, in the process of obtaining the ratio of interaction energy between two isotopes, we are comparing very similar spectra, and therefore the effects due to the neglected factors tend to be canceled when the ratio is taken. Strengthening this argument is the fact that, in Table II, the ratios of interaction energy for Gd^{160} compared to Gd^{155} are very consistent within statistical error when three different forms of absorber are used. This is in contrast to the observation in Table I that the absolute value of quadrupole splitting in Gd^{155} varies more widely than the statistical

TABLE II. Comparison of quadrupole moments for the even-even isotopes of gadolinium.[a]

Isotope	Absorber	$E_Q{}^A/E_Q{}^{155}$	$Q_0{}^A/Q_0{}^{155}$	Q^A/Q^{155} from Coulomb-excitation cross section (Ref. 26)
Gd^{156}	Gd_2O_3	0.76±0.03	1.07±0.04	1.04±0.02
Gd^{158}	Gd_2O_3	0.82±0.02	1.15±0.03	1.14±0.02
Gd^{160}	Gd_2O_3	0.84±0.02	1.17±0.03	1.18±0.02
	Metallic Gd	0.86±0.04	1.21±0.05	
	$GdAl_2$	0.82±0.03	1.15±0.03	

[a] Uncertainty for Mössbauer data is statistical on the basis of least-squares fit to the assumed hyperfine-interaction forms, which is reliable when ratios of interaction energies or ratios of moments are concerned. See the text for discussion of this point.

errors indicate, depending on the form of absorber. For each case of Gd^{160} absorber in Table II, we use the value of $E_Q{}^{155}$ corresponding to the same form of absorber as presented in Table I.

E. Comparison of Quadrupole Moments of the Gadolinium Isotopes

The quadrupole-interaction energies extracted in the even-even isotopes of gadolinium can be compared to the quadrupole-coupling energies observed with the same absorbers, using the $Eu^{155}{}_2O_3$ source, and ratios of quadrupole moments thereby obtained. In making these comparisons, it is necessary to take into account the projection factors which relate the observed quadrupole moments Q_I to the intrinsic quadrupole moments Q_0. Using the results of Bohr,[25] we have

$$Q_0{}^A=\frac{(I+1)(2I+3)}{3K^2-I(I+1)}Q_I$$

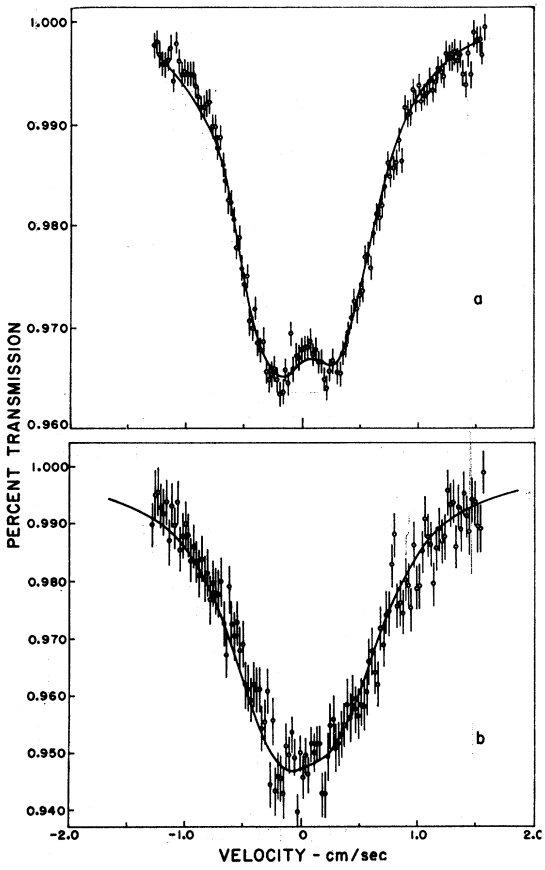

FIG. 9. Mössbauer absorption spectra following Coulomb excitation of the 75.3-keV state of Gd^{160} at 4.2°K for (a) $GdAl_2$ absorber containing 21 mg/cm² of Gd^{160} and (b) metallic Gd absorber containing 46 mg/cm² of Gd^{160}. These data have been corrected for nonresonant background.

[25] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953).

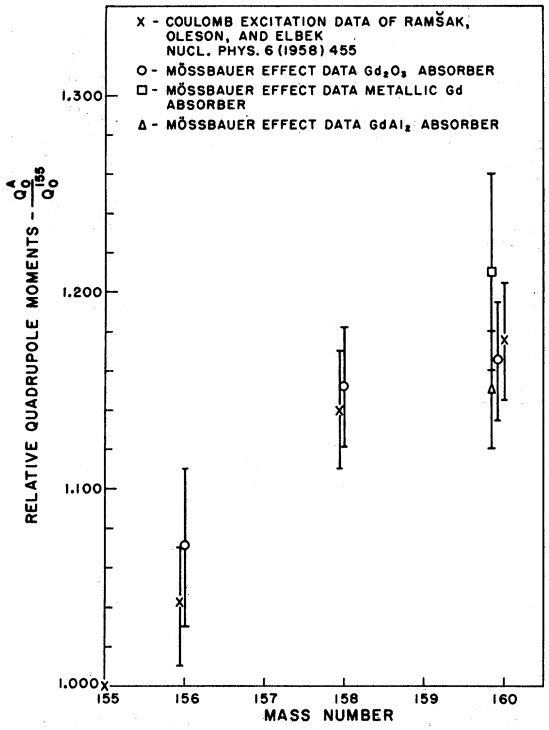

FIG. 10. Relative quadrupole moments of even-even isotopes of gadolinium.

and we obtain

$$\frac{Q_0{}^A}{Q_0{}^{155}}=-\frac{7}{5}\frac{E_Q{}^A}{E_Q{}^{155}}.$$

Only magnitudes of quadrupole moments have been considered here, as the present data are not sufficient to determine the relative sign of $Q_0{}^{155}$ and $Q_0{}^A$.

The ratios of quadrupole moments $Q_0{}^A/Q_0{}^{155}$ are presented in Fig. 10, together with corresponding results obtained from Coulomb-excitation cross-section data.[26] The two determinations are seen to agree within the experimental uncertainties, and thus these measurements provide additional confirmation of the collective theory of deformed nuclei. The absolute values of Q_0 for the Mössbauer data can be obtained by using the value of $Q_0{}^{155}$ obtained from spectroscopic data,[27] which agrees with the value obtained from Coulomb-excitation data.

F. Dy^{164} Results Following Coulomb Excitation

Well-resolved magnetic hyperfine structure was observed in the Mössbauer absorption spectrum of $Dy^{164}{}_2O_3$ following Coulomb excitation of the 73.4-keV

[26] V. Ramsak, M. C. Olesen, and B. Elbek, Nucl. Phys. **6**, 451 (1958).

[27] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, D. C.), NCR 5-5-59.

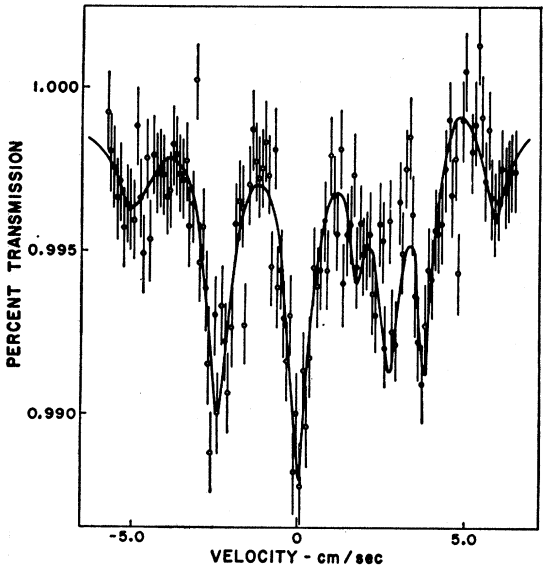

FIG. 11. Mössbauer absorption spectrum following Coulomb excitation of the 73.4-keV state of Dy^{164} at 4.2°K for a Dy_2O_3 absorber containing 20 mg/cm² of Dy^{164}. These data have been corrected for nonresonant background.

state of Dy^{164}. An enriched sesquioxide target was bombarded with a 0.2-μA beam of 3-MeV protons; the γ-ray peak following Coulomb excitation was well resolved from the K x rays, and a signal-to-noise ratio of 4:1 was observed with a counting rate of 990 counts/sec in the γ-ray window. A natural Dy_2O_3 absorber was used which contained 20 mg/cm² of Dy^{164}.

In previous work Cohen[28] has established that Dy_2O_3 exhibits large magnetic splitting by using a $TbAl_2$ source with an enriched $Dy^{160}{}_2O_3$ absorber and observing the Mössbauer effect in Dy^{160}. Assuming pure magnetic splitting, we would expect a split-split pattern of nine equally spaced absorption lines. The absorber velocity range was set at ± 6 cm/sec which, assuming a comparable spectrum to that observed by Cohen, should encompass five of these nine lines. The observed spectrum is shown in Fig. 11. Additional structure besides the expected five lines was observed in the positive portion of the spectrum, which could be due to radiation damage in the target, although positive identification of this structure was not possible. The uncertainty in the interpretation of the split-split pattern precludes any definite conclusion as to the relative gyromagnetic ratio, but such a determination should be possible for all the even-even dysprosium isotopes if an effectively monochromatic source can be obtained.

G. Er^{168} Results Following Coulomb Excitation

Mössbauer studies were carried out in Er^{168} following Coulomb excitation of the 79.8-keV state using both oxide and metallic absorbers. In both cases enriched sesquioxide targets of $Er^{168}{}_2O_3$ were bombarded with a 0.2-μA beam of 3-MeV protons. The Coulomb-excited γ ray was partially masked by the K x rays of erbium. Counting rates of 700 counts/sec were obtained, with a signal-to-noise ratio of 1:1 with the oxide absorber, and 3:1 with the metallic absorber. The spectra observed are presented in Fig. 12.

In previous work Cohen *et al.*[29] have shown that the Mössbauer absorption spectrum for Er_2O_3 exhibited a single absorption line by observing the Mössbauer effect in Er^{166} using an effectively unsplit source of $HoAl_2$. The present data with the oxide absorber exhibit a somewhat broader line, since both an oxide target and an absorber were used. There is an additional absorption dip at 2.8 cm/sec in the Coulomb-excitation data; it is hypothesized that this peak may be a radiation-damage peak.

The spectrum obtained with the metallic-erbium absorber exhibits the expected five-line absorption pattern anticipated previously from work with Er^{166}

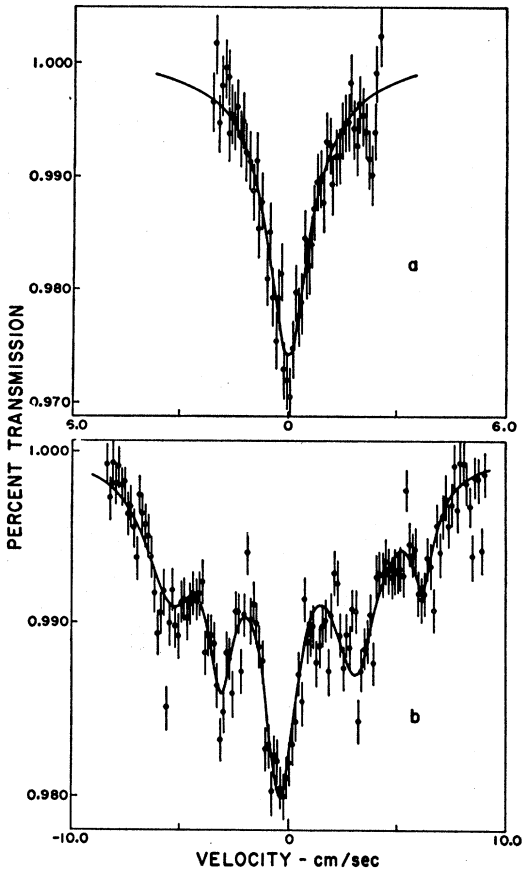

FIG. 12. Mössbauer absorption spectra following Coulomb excitation of the 79.8-keV state of Er^{168} at 4.2°K for (a) Er_2O_3 absorber containing 62 mg/cm² of Er^{168} and (b) metallic erbium absorber containing 31 mg/cm of Er^{168}. These data have been corrected for nonresonant background.

[28] R. L. Cohen, Phys. Rev. **137**, A1809 (1965).

[29] R. L. Cohen and J. H. Wernick, Phys. Rev. **134**, B503 (1964).

by Kienle.[30] This spectrum exhibits very broad absorption lines which are due in part, to the linewidth of the target. Assuming that the structure was due to magnetic hyperfine interaction with a small admixture of quadrupole splitting, a magnetic energy splitting of $(7.8\pm0.4)\times10^{-6}$ eV and a quadrupole-interaction energy of $(0.9\pm0.4)\times10^{-6}$ eV were obtained. Using the effective magnetic field in erbium of $H=7.46\times10^{6}$ Oe,[31] we obtain a value of g_R of 0.33 ± 0.02. This result agrees within the experimental error with the value of 0.31 predicted by Nilsson and Prior.[32] Further work with a target having a narrower linewidth should permit a more accurate determination of the values of g_R for all the even-even isotopes of erbium.[33]

[30] P. Kienle, Rev. Mod. Phys. **36**, 372 (1964).
[31] H. Dobler, G. Petrich, S. Hüfner, P. Kienle, W. Wiedemann, and H. Eicher, Phys. Letters **10**, 319 (1964).
[32] S. G. Nilsson and O. Prior, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **32**, No. 16 (1961).
[33] After the publication of the present result by J. Eck, Y. K. Lee, E. T. Ritter, R. R. Stevens, Jr., and J. C. Walker [Phys. Rev. Letters **17**, 120 (1966)] there was another experiment by E. Münk, D. Quitman, and S. Hüfner [Z. Naturforsch **21**, 847 (1966)] who reported $g_R=0.331\pm0.010$ for Er^{168}, in good agreement with the present results.

V. CONCLUSIONS

Mössbauer studies were carried out following Coulomb excitation in Gd^{155}, Gd^{156}, Gd^{158}, Gd^{160}, Dy^{164}, and Er^{168}. For the last three isotopes there are no suitable radioactive parents, and the present work is the first observation of Mössbauer effect in these nuclides. The systematic variation of the quadrupole moments in Gd^{156}, Gd^{158}, and Gd^{160} was observed by using Coulomb excitation to populate the first excited states of these nuclei, and subsequently observing the Mössbauer effect from these levels. The ratios of quadrupole moments obtained agreed within the experimental uncertainty with the corresponding ratios obtained from Coulomb-excitation cross-section data, and thus these results provide additional confirmation of the collective theory of deformed nuclei.

The results of these experiments demonstrate the feasibility of carrying out systematic studies of nuclear properties using the Mössbauer effect following Coulomb excitation, and further demonstrate that the highly deformed nuclei in the rare earths are quite accessible to detailed study by this technique.

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Fission and the Synthesis of Heavy Nuclei by Rapid Neutron Capture*

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The role of fission is examined in the synthesis of heavy nuclei by multiple capture of neutrons in thermonuclear explosions. We begin by reviewing evidence from the recent Tweed and Cyclamen experiments indicating that neutron-induced fission is a serious source of depletion in neutron capture chains which start from targets of ^{242}Pu and ^{243}Am. An analysis of Tweed abundances (Sec. 2) is made to obtain capture-to-fission ratios for the odd-A plutonium isotopes through $A=253$. We next use the liquid-drop model of Myers and Swiatecki plus empirical shell corrections and pairing energies, in order to correlate and predict spontaneous fission lifetimes (Sec. 3) and fission barriers (Sec. 4). For nuclei having $Z\leq101$ and $N\leq157$, we extrapolate the shell correction, assuming it to be a function of N plus a function of Z, and thus obtain neutron binding energies, fission barriers, and spontaneous fission lifetimes for neutron-rich heavy nuclei (Sec. 6). Capture-to-fission ratios are estimated for many of these nuclei in Sec. 7, and qualitative agreement is found with laboratory and Tweed results. In Sec. 8, the extrapolation is continued out to $N=159$ and $Z=104$. We conclude that by using the liquid-drop model plus semiempirical shell corrections, one can obtain capture-to-fission ratios and spontaneous fission half-lives which are usefully accurate. However, for predicting properties of nuclei having $Z>104$, $N\gtrsim159$, one needs, in this formalism, an accurate way of predicting shell corrections or nuclear masses.

1. INTRODUCTION

WE consider the synthesis of heavy nuclei by the multiple capture of neutrons in nuclear explosions. In debris from the first experiment of this type, namely the Mike thermonuclear event of November 1952, nuclei through mass 255 were detected,[1] starting from a target of ^{238}U. In two more recent and more readily interpretable experiments, the Par[2] and Barbel[3] underground explosions of October 1964, nuclides through mass 257 were identified, again starting from a target of ^{238}U. A theoretical interpretation of the abundances of the various neutron capture products

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] H. Diamond *et al.*, Phys. Rev. **19**, 2000 (1960).
[2] D. W. Dorn and R. W. Hoff, Phys. Rev. Letters **14**, 440 (1965).
[3] Los Alamos Radiochemistry Group, Phys. Rev. Letters **14**, 962 (1965).