Ratio of the Quadrupole Moment of the First Excited State of Xe^{129} to That of the Ground State of Xe^{131}

GILBERT J. PERLOW*

Argonne National Laboratory, Argonne Illinois

(Received 17 April 1964)

The Mössbauer effect in Xe^{129} and Xe^{131} contained in XeF_4 has been used to measure the ratio of electric quadrupole moments of the lowest $\frac{3}{2}^+$ states in the two isotopes. This is the ground state in Xe^{131} and the first excited state in Xe^{129}. The value $Q(Xe^{129*})/Q(Xe^{131})=3.45\pm0.09$ is obtained. The sign of the moment in Xe^{129} is obtained by use of a source containing oriented crystals of $KI^{129}Cl_4$ which produces $XeCl_4$ in its beta decay. The quadrupole moment is found to be negative. The known quadrupole moment of the ground state of Xe^{131}, -0.12b, then gives $Q(Xe^{129*}) = -0.41$b. The large ratio of the moments is interpreted as due to the abrupt onset of a region of permanent deformation by analogy with a similar situation in the europium isotopes. Discussions of such a region appear in the literature. The linewidth observed in Xe^{131} yields a value for the first-excited-state lifetime, $T_{\frac{1}{2}}=0.504\pm0.017$ nsec, in good agreement with delayed-coincidence measurements. The measured value for the ratio of the moments removes a difficulty in understanding the structure of xenon fluorides generated by an earlier assumption that the ratio was unity.

INTRODUCTION

THE Mössbauer effect in $Xe^{129}F_4$ and $Xe^{131}F_4$ has been used to obtain the ratio of quadrupole moments of the lowest $\frac{3}{2}^+$ states in the two xenon isotopes. In Xe^{129}, the ground state is $\frac{1}{2}^+$ and the first excited state at 39.6 keV is $\frac{3}{2}^+$. Addition of two neutrons appears merely to invert the level order: Xe^{131} has a $\frac{3}{2}^+$ ground state and a $\frac{1}{2}^+$ first excited state at 80.2 keV. Shortly after the synthesis[1] of XeF_4, we observed[2,3] quadrupole splitting in the Mössbauer velocity spectrum of $Xe^{129}F_4$ and $Xe^{129}F_2$, the source being 1.6×10^6 yr I^{129} in the form of NaI. In order to analyze the molecular structure of these substances from the splitting, the quadrupole moments of the $\frac{3}{2}^+$ states in the two nuclei were assumed to be identical. The value of $Q(Xe^{131})$ was known from optical[4] and atomic-beam[5] measurements. The assumption of equality appeared justified on various grounds. In particular, of 33 equivalent cases found in the literature, a ratio of moments (or intrinsic moments for $J=0$) larger than 1.6 occurred only once. This was the case $Q(Eu^{153})/Q(Eu^{151})=2.26$.[6] It is known that the europium isotopes lie on either side of the lower edge of a region of permanent deformation.[7,8] With the moments assumed equal, the splitting in the xenon fluorides could not readily be understood. Initial efforts to measure $Q(Xe^{129*})/Q(Xe^{131})$ with various I^{131} sources in the form

of iodides were unsuccessful; but the attempts led to the discovery that xenon compounds could be formed as a result of the beta decay of iodine compounds having high formal valence.[9] The xenon is then relatively tightly bound in the lattice and has a recoilless fraction 2 to 3 times as great as that observed with an iodide source. Sources of KIO_4 and $Na_2H_3IO_6$ containing I^{129} were made and were found to emit single lines when used with an unsplit absorber. With $Na_2H_3IO_6$ made with I^{131}, we have been able to see the Mössbauer effect in $Xe^{131}F_4$ and, by comparison with $Xe^{129}F_4$, to obtain the ratio of quadrupole moments. The Xe^{129*} is found to have a quadrupole moment 3.45 times as large as that of Xe^{131}. It is natural to assume that, just as in the case of the Eu isotopes, the two straddle the edge of a region of permanent deformation. Experiments have also been done to determine the sign of the quadrupole moment in Xe^{129*}. It is found to be negative as in Xe^{131}.

APPARATUS

The velocity spectrometer used was a mechanical refinement of that described in Refs. 2, 3, and 6. The absorber was mounted in a yoke extending below the lower end of a vertical stainless-steel tube which reached down into a glass liquid-helium Dewar. At its upper end the tube was held in a parallelogram mount and driven up and down by a crank on a flywheel. The amplitude of motion was adjustable and was about 1 mm for most of the runs. A stationary source was fastened above the absorber.

The detector, located below the Dewar, was a proportional counter for the 40-keV radiation or a NaI scintillator for the 80 keV. Pulses were selected in a single-channel analyzer whose output was stored in successive memory locations of a 400-channel analyzer used as a multiple scaler. The memory address was advanced by a scaled-down crystal oscillator which was reset and

* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] H. H. Claassen, H. Selig, and J. G. Malm, J. Am. Chem. Soc. 84, 3593(1962).

[2] C. L. Chernick, C. E. Johnson, J. G. Malm, G. J. Perlow, and M. R. Perlow, Phys. Letters 5, 103 (1963).

[3] G. J. Perlow, C. E. Johnson, and M. R. Perlow, in *Noble Gas Compounds*, edited by H. Hyman (University of Chicago Press, Chicago, 1963), p. 279.

[4] A. Bohr, J. Koch, and E. Rasmussen, Arkiv Fysik 4, 455 (1952).

[5] W. L. Faust and M. N. McDermott, Phys. Rev. 123, 198 (1961).

[6] M. Abraham, R. Kedzie, and C. D. Jeffries, Phys. Rev. 108, 58 (1957).

[7] K. Ford, Phys. Rev. 90, 29 (1953).

[8] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Skrifter 1, No. 8 (1959).

[9] G. J. Perlow and M. R. Perlow, Rev. Mod. Phys. 36, 353 (1964).

restarted by the action of a shutter on a light beam. The shutter was carried by the flywheel which was driven by O rings from a synchronous motor. The velocity of the system was calibrated by measuring the adjustable crank throw, the flywheel frequency, and the frequency of address advance. The channel number corresponding to zero velocity was determined by an auxiliary measurement based on the Mössbauer effect in Fe[57]. In our method of operation, the velocity is a sinusoidal function of channel number and requires converting. The count scale is uniform. The velocity is known within about 0.2%. The channels are equal in width (at least statistically) to considerably better than 0.01%, as judged by the values of χ^2 obtained in runs with about 2×10^7 counts per channel.

Xenon tetrafluoride decomposes if exposed to air. The sample, 1.66 g of the compound made with Xe in its natural abundance, was contained in a flat-sided quartz cell 1 in. in diameter. The absorber was prepared for each run by subliming the material in the cell upon one flat surface by means of a temperature gradient normal to the surface.

The least-squares treatment of the data followed the variable-metric minimization technique which was developed by Davidon[10] and was applied to the problem of fitting Mössbauer spectra by M. Welch, B. S. Garbow, and J. P. Schiffer. The absorption peaks were fitted with Lorentzians. Data from several runs were averaged with due regard to the errors assigned to the parameters by the fitting program.

All of the experiments reported on here were done with both source and absorber at 4.2°K.

DATA

Figure 1 shows representative velocity spectra with the same XeF$_4$ absorber and separate unsplit sources of I[129] and I[131]. The curves are plotted to the same energy scale. The lower curve is the sum of two runs. The ratio
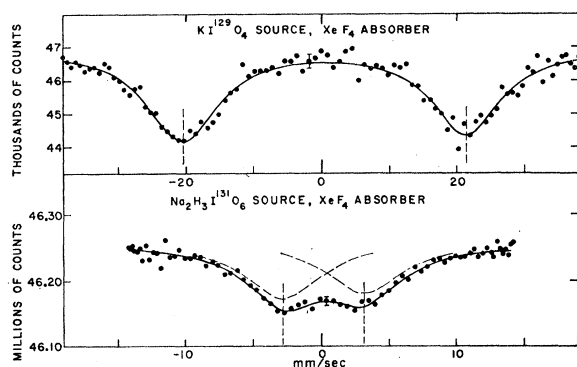
of quadrupole moments is given directly by the ratio of the splitting in energy units. The decomposition of the doublet structure in Xe[131]F$_4$ into component Lorentzians is shown by the dashed curves. All of the seven independent runs on Xe[131] with various velocity dispersions had acceptable values of χ^2. Table I shows the results.

TABLE I. Summary of data and conclusions.

Absorber	Splitting (mm/sec)	e^2qQ (Mc/sec)	Linewidth (mm/sec)
Xe[129]F$_4$	41.74±0.29	2666±19	12.38±0.5
Xe[131]F$_4$	5.97±0.16	772±21	6.76±0.23

$$|Q(\text{Xe}^{129*})/Q(\text{Xe}^{131})| = 3.45 \pm 0.09$$
$$Q(\text{Xe}^{129*}) = -0.41 \pm 15\%^a$$
$$\text{Deformation } \delta = -0.14$$

[a] Based on $Q(\text{Xe}^{131}) = -0.12 \pm 15\%$, Ref. 4.

The final value of the ratio is

$$Q(\text{Xe}^{129*})/Q(\text{Xe}^{131}) = 3.45 \pm 0.09.$$

The error quoted is the statistical one.

The linewidth gives a lower limit to the lifetime of the state. For Xe[129], we get $T_{1/2} = 0.56 \pm 0.02$ nsec, as compared with 1.01 ± 0.04 nsec by delayed coincidence.[11] For Xe[131] we get 0.504 ± 0.017 nsec, in excellent agreement with the value[12] 0.496 ± 0.021 nsec obtained by delayed coincidence.

For Xe[131] the absorber was effectively quite thin, while it was thick for Xe[129]. If the broadening of the lines in the Xe[129] measurement is ascribed to absorber thickness, we find that $fn\sigma_0 = 6.5$, where f is the recoilless fraction in the absorber, n is the thickness (atoms/cm^2) of Xe[129], and σ_0 is the peak cross section. If $\alpha = 9$ is taken as the internal-conversion coefficient for the 40-keV state, then $\sigma_0 = 0.15 \times 10^6$b and $f = 0.22$ at liquid-helium temperature. This leads to a value $\theta_D = 78°$ for the Debye temperature. This does not purport to be a good measurement of θ_D, but the value is reasonable and lends support to the idea that the line broadening in Xe[129]F$_4$ is due in considerable part to absorber thickness.

For the case of spin $\frac{3}{2}$, the splitting is related to the electric quadrupole moment Q and the electric-field gradient $-eq$ by the expression $\Delta E = e^2qQ/2$ when, as in XeF$_4$, there is a fourfold symmetry axis. Conversion from mm/sec to Mc/sec may be made by multiplying the former by 31.94 for Xe[129*] and by 64.66 for Xe[131]. The process of extracting quadrupole moments from couplings involves the well-known shielding uncertainty.[13] We use the uncorrected value $Q = -0.12$b of Ref. 4, wherein it is estimated that the correction is less than 5% and the entire error less than about 15%.

FIG. 1. Velocity spectra in Xe[129] and Xe[131]. The velocity scales have been adjusted so that the energy scales are approximately equal.

[10] W. R. Davidon, Argonne National Laboratory Report No. ANL-5990, 1959 (unpublished).

[11] J. S. Geiger, R. L. Graham, F. Brown, and I. Bergstrom, Phys. Can. 18, No. 3, 21 (1962).
[12] R. S. Weaver, Can. J. Phys. 40, 1684 (1962).
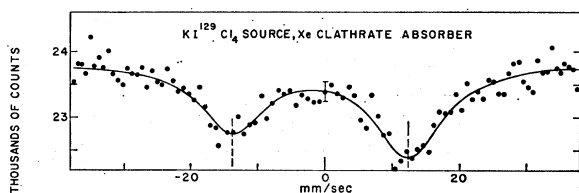[13] R. Sternheimer, Phys. Rev. 80, 102 (1950).

FIG. 2. Velocity spectrum of the 40-keV γ radiation emitted from $Xe^{129}Cl_4$ produced in the β decay of oriented crystals of $KI^{129}Cl_4$.

From this, and anticipating the result of the following section,

$$Q(Xe^{129*}) = -0.41b.$$

Determination of the Sign of Q(Xe^{129*})

The sign of the quadrupole moment may be determined by an angular-distribution measurement on oriented molecules. If the molecular symmetry axis is taken as z axis, a gamma-ray transition between $J=\frac{3}{2}$ and $J=\frac{1}{2}$ has an intensity distribution proportional to $1\pm\frac{1}{2}P_2(\cos\theta)$. The positive sign applies to the multiplet component characterized by $|m_z|=\frac{3}{2}\leftrightarrow|m_z|=\frac{1}{2}$, and the negative sign to $|m_z|=\frac{1}{2}\leftrightarrow|m_z|=\frac{1}{2}$. The intensities are equal for a randomly oriented powder where $\langle P(\cos\theta)\rangle_{av}=0$, or for an aligned molecule observed at $54°44'$.

Efforts to work in absorption with aligned samples of XeF_4 sufficiently large in size and appropriately shaped were of only marginal success. It was found possible, however, to do the measurement in emission with another substance. We have submitted elsewhere a report on the production and electronic structure of $XeCl_4$ produced by the beta decay of $I^{129}Cl_4^-$. This case is more favorable to a determination of the sign by orientation. The crystal $KICl_4\cdot H_2O$ grows in needles in which the square planar ICl_4^- molecular ions (symmetry group D_{4h}) have their fourfold axes tilted at about 30° to the needle axis.[14] A source of such crystals containing I^{129} was prepared and oriented perpendicular to the direction of observation. The molecular symmetry axes then formed a cone of apex angle $2\alpha\doteq60°$ whose axis was perpendicular to the direction of observation. Under these circumstances, one expects the intensities of the multiplet components to be

$$\frac{I(\pm\frac{1}{2}\to\pm\frac{1}{2})}{I(\pm\frac{3}{2}\to\pm\frac{1}{2})}=\frac{\frac{5}{3}-\frac{1}{2}\sin^2\alpha}{1+\frac{1}{2}\sin^2\alpha}=1.4.$$

Figure 2 shows an experimental spectrum. The absorber was the hydroquinone clathrate of xenon containing 37 mg/cm² of Xe. This has been shown[2] to be unsplit. From 3 runs, the ratio of areas is found by the curve-fitting routine to be 1.37 ± 0.11, in agreement with expectation. The peak at positive velocity (i.e., with source approaching absorber) is the more intense and

indicates that the state $|m|=\frac{1}{2}$ is the lower, and hence that the quadrupole coupling e^2qQ is positive.

The substance $KICl_4\cdot H_2O$ has been thoroughly studied by the technique of nuclear quadrupole resonance.[15] The bonding of the iodine results in a local excess of negative charge above and below the molecular plane. That is $eq\equiv\partial^2V/\partial z^2<0$. From the similarity of the Mössbauer patterns of $XeCl_4$ and XeF_4, and from the often remarked[16,17] similarity of square planar XeF_4 to ICl_4^- [a similarity strongly reinforced by the present Mössbauer results on the magnitude of $Q(Xe^{129*})$], it is regarded as a quite reasonable assumption that the bonding in $XeCl_4$ results also in $q<0$. It then follows that $Q(Xe^{129*})<0$.

DISCUSSION

For a uniformly charged spheroidal nucleus with deformation parameter δ, the quadrupole moment Q_0 with respect to the body-fixed axes is given by[8]

$$Q_0=\tfrac{4}{5}\delta ZR_0^2(1+\tfrac{1}{2}\delta+\cdots).$$

The measurable moment Q for spin $\frac{3}{2}$ and strong coupling is[18]

$$Q=\pm\tfrac{1}{5}Q_0.$$

The positive sign is to be used if, as seems most reasonable (and as we shall assume), the $\frac{3}{2}^+$ state is intrinsic; the negative sign if it is to be considered a member of rotation band based on the $\frac{1}{2}^+$ ground state. The value for $Q(Xe^{129*})$ leads to the value $\delta=-0.14$ if $R_0=1.2\times A^{1/3}\times10^{-13}$ cm. The deformation parameters in the rare-earth region tend to be about three times as large. Ford[7] analyzed quadrupole-moment data which suggested a region of permanent deformation for Z and N between 50 and 82. Sheline and collaborators,[19] who measured the energies of the first excited states of Ba^{126}, Ba^{128}, and Ba^{130}, found indications of a deforming trend. They also calculated[20] the expected deformation in this region by the method of Mottelson and Nilsson.[21] Pairing interactions for the region have been included in a more recent calculation by Kumar and Baranger,[22] who also concluded that there should be deformation and, more specifically, that it should be of oblate character ($\delta<0$). The deformation observed in the present experiment is substantially equal in magnitude to that predicted in Ref. 20

It may be remarked in passing that the value ob-

[14] R. C. L. Mooney, Z. Krist. 98, 377 (1938).

[15] C. D. Cornwell and R. S. Yamasaki, J. Chem. Phys. 27, 1060 (1957); R. S. Yamasaki and C. D. Cornwell, ibid. 30, 1265 (1959).
[16] K. S. Pitzer, Science 139, 414 (1963).
[17] R. E. Rundle, J. Am. Chem. Soc. 85, 112 (1963).
[18] A. K. Kerman, in Nuclear Reactions, edited by P. M. Endt and M. Demeur (North-Holland Publishing Company, Amsterdam, 1959), Vol. 1, p. 440.
[19] R. K. Sheline, T. Sikkeland, and R. N. Chanda, Phys. Rev. Letters 7, 446 (1961).
[20] E. Marshalek, L. W. Person, and R. K. Sheline, Rev. Mod. Phys. 35, 108 (1963).
[21] B. R. Mottelson and S. G. Nilsson, Phys. Rev. 99 1615 (1955).
[22] K. Kumar and M. Baranger, Phys. Rev. Letters 12, 73 (1964).

tained here for $Q(\text{Xe}^{129*})$ brings the Mössbauer-effect results on the structure of xenon fluorides into agreement with expectations[23] based on other types of measurement.

ACKNOWLEDGMENTS

The author would like to acknowledge helpful conversations at various times with A. Arima, S. Ruby,

²³ D. Lazdins, C. W. Kern, and M. Karplus, J. Chem. Phys. 39, 1611 (1963).

Recoil Studies of Nuclear Reactions Induced by High-Energy Particles. I. Production of Tb¹⁴⁹†

LESTER WINSBERG*

Argonne National Laboratory, Argonne, Illinois
and
Lawrence Radiation Laboratory, University of California, Berkeley, California

(Received 17 February 1964)

The interaction of high-energy protons and alpha particles with Ta, Au, and Bi to produce Tb¹⁴⁹ was investigated by the recoil technique and by measuring the excitation functions. The experimental results were analyzed in terms of a two-step mechanism: (1) An initial interaction causes the struck nucleus to recoil forward. (2) This excited nucleus then loses its energy of excitation by emitting various particles until the final nucleus is formed. The results of this analysis confirm the main features of this mechanism. These results were further analyzed for the details of the mechanism by applying the laws of energy and momentum conservation in a general way.

IT is almost axiomatic that one gains the greatest insight into any process by studying it from many points of view. This is particularly true of nuclear reactions where the phenomena are complex and the experimental results difficult to interpret. On the other hand, the number of different types of experiment one can perform in any given investigation are limited. The study reported here was initiated in order to see how much could be learned by applying the simplest techniques to a typical nuclear reaction.

The reaction studied was the production of Tb¹⁴⁹ from Ta¹⁸¹, Au¹⁹⁷, and Bi²⁰⁹. Here, the product is appreciably lighter than the target nucleus. The reactions were initiated by protons and alpha particles with energies above 400 MeV. The maximum proton energy was 6.2 BeV; for alpha particles, 0.88 BeV.

These systems were chosen because the target foils are readily available, and because the ground state of Tb¹⁴⁹ is an easily identifiable alpha-particle emitter.[1–3]

The target foils were exposed to proton beams from the bevatron in Berkeley and the Fermi Institute cyclotron at the University of Chicago and to protons and alpha particles from the 184-in. cyclotron in Berkeley. The excitation functions and recoil properties of the final product, Tb¹⁴⁹, were measured.

Thus, we have two types of measurement made over a range of bombarding energies with both protons and alpha particles. This variety of information was essential for revealing some of the salient features of the reaction mechanism given in the Summary.

EXPERIMENTAL PROCEDURE

Two types of experiments were performed: In the first type the target foil [No. 1 in Fig. 1(a)] and the Al catcher foils surrounding them (No. 2) were thick (several mg/cm²) relative to the range of the recoiling Tb¹⁴⁹. Several additional ¼-mil Al foils were included as cross-section monitors (No. 3). Aluminum cover foils (No. 4) protected the ensemble. The excitation functions are based primarily on data obtained in these experiments. Values of the cross sections obtained in the second type of experiment agree with these values to better than 10%.

In the second type of experiment the targets consisted of thin films (mostly of thickness ~30 μg/cm²) of

¹ J. O. Rasmussen, S. G. Thompson, and A. Ghiorso, Phys. Rev. 89, 33 (1953).
² L. Winsberg, Bull. Am. Phys. Soc. 3, 406 (1958).
³ R. D. Macfarlane, Phys. Rev. 126, 274 (1962).