Isotope Shift in the Muonic K X Rays of O^{16}-O^{18}, Ca^{40}-Ca^{44}, Sn^{116}-Sn^{124}, and Gd^{155}-Gd^{160}†

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Measurements have been made of the isotope shift in the muonic K x rays for the pairs of isotopes O^{16}–O^{18}, Ca^{40}–Ca^{44}, Sn^{116}–Sn^{124}, and Gd^{155}–Gd^{160}. The measurements were limited in resolution by the conventional NaI detectors used: no fine or hyperfine structure was resolved. The results confirm the indications from optical isotope-shift measurements that the expansion in charge distribution of a nucleus on addition of neutrons is less than the expansion in the density distribution, and indicate that this trend is even more marked in the light nuclei examined.

I. INTRODUCTION

EXTENSIVE investigations of the radial distribution of electric charge in atomic nuclei have been made both by high-energy electron scattering experiments[1] and by measurement of the $2p$-$1s$ transitions in muonic atoms[2]; but these investigations have been almost exclusively limited to nuclei with the neutron-proton ratio corresponding to maximum stability, especially those cases where a particular element is represented wholly or predominantly by a single naturally occuring isotope. The manner in which the charge distribution depends on neutron number for isotopes of a given element has also been studied by measurement of the so-called "isotopic shift" in atomic optical spectra,[3] but these measurements have been restricted to heavier nuclei ($A \gtrsim 100$) and their full interpretation is limited by imprecise knowledge of the relevant electron wave functions. Precision measurements of the theoretically much simpler muonic spectra do, in principle, afford a means of extending knowledge of the dependence of nuclear charge distribution on neutron number, but the effects to be measured are extremely small. With conventional techniques it is quite impossible, for example, to resolve the spectral lines relating the different isotopic components that would be obtained from materials containing several isotopes with their normal abundances. However, by using the relatively large quantities (10–100 g) of separated isotopes now available, the *difference* between one isotope and another of the energies of the prominent lines (especially $2p$-$1s$ transitions) in the muonic spectra can be determined with moderate precision. Although such measurements are, as will be seen, not of the same precision as the optical isotope shift measurements, their interpretation is so much more direct that the two types of measurement can be regarded as largely complementary. A relatively small number of muonic measurements could enhance the value of the optical isotopic shift measurements now available.

In the following we describe the measurements of energy differences for the $2p$-$1s$ muonic x-ray transitions in the following pairs of isotopes: O^{16}-O^{18}, Ca^{40}-Ca^{44}, Sn^{116}-Sn^{124}, and Gd^{155}-Gd^{160}, using a conventional NaI scintillator as detector.[4] In such low-resolution measurements none of the finer details, the $2p_{1/2}$-$2p_{3/2}$ splitting, the quadrupole hyperfine interactions, etc., of the spectra are resolved. For the spinless spherical nuclei, O, Ca, and Sn, this limitation does not probably materially influence the interpretation; for the gadolinium isotopes the situation is more complex as discussed below. In all cases the expected isotope shift is very small (and the measured shift was smaller still) less than one-tenth of the experimental linewidths. The measurement of such small energy differences is only possible by exploiting the high detection efficiency of the NaI detector and by taking the most scrupulous measures to avoid drifts in the detector energy sensitivity, and other sources of error which are not statistically random fluctuations.

II. EXPERIMENTAL

The critical feature of our experimental procedure was the essentially simultaneous measurement of the muonic x rays from a pair of isotopes in an arrangement which had identical sensitivity for both spectra. The negative muon beam from the Columbia University Nevis synchrocyclotron was produced and slowed down in the usual way. An improved "duty cycle" was obtained by use of a vibrating target[5]; muons were produced during one-third of the accelerator cycle of 16.7 msec. The beam intensity was quite uniform during the "on" time except for short bursts at the beginning and end of each cycle. In order to reduce accidental coincidences, the muon detection apparatus was sensitive only during the period of uniform beam intensity.

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[1] R. Hofstadter, Ann. Rev. Nucl. Sci. **7**, 231 (1957).
[2] V. L. Fitch and J. Rainwater, Phys. Rev. **92**, 789 (1953); P. Brix, R. Engler, U. Hegel, D. Quittmann, G. Backenstoss, K. Goebel, and B. Stadler, Phys. Letters **1**, 56 (1962); C. S. Johnson, E. P. Hincks, and H. L. Anderson, Phys. Rev. **125**, 2102 (1962); **130**, 2468 (1963); and W. Frati and J. Rainwater, *ibid.*, **128**, 2360 (1962).
[3] G. Breit, Rev. Mod. Phys. **30**, 507 (1958); and P. Brix and H. Kopfermann, *ibid.* **30**, 517 (1958).
[4] A preliminary account of the calcium measurements has been published: R. Cohen, S. Devons, A. Kanaris, and C. Nissim-Sabat, Phys. Letters **11**, 70 (1964).
[5] J. L. Rosen, Nevis Report No. 92, Columbia University, 1960 (unpublished).

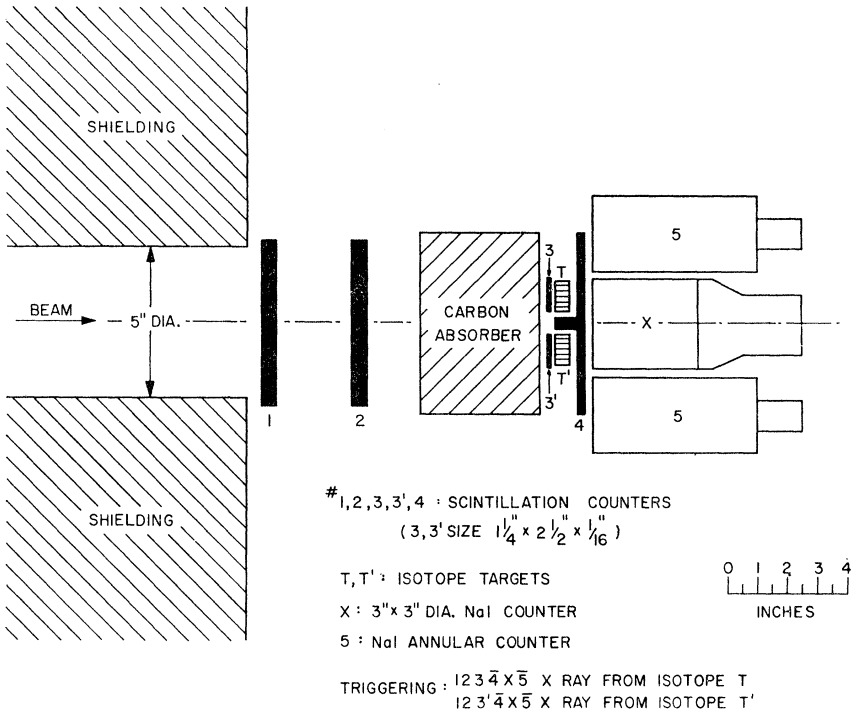

FIG. 1. Schematic diagram of the counter telescope and of the target and NaI crystal arrangement.

After passing through plastic scintillator counters Nos. 1 and 2, and some 4 in. of graphite moderator (Fig. 1), the muons impinged on a pair of thin scintillation counters Nos. 3 and 3′ placed side by side, behind which were placed the two isotope samples T and T' which formed the stopping target. The two targets T and T' were alike in every respect other than isotopic number. The thin scintillator counter No. 4 placed behind this composite target was used in anticoincidence and indicated a stopping muon; the coincidence circuits $123\bar{4}$, $123'\bar{4}$ indicated that a muon had stopped in target T and T', respectively. (The anticoincidence counter No. 4 was shaped so as to eliminate from consideration muons entering one-half of Nos. 3 and 3′ and after being scattered in this counter or the target material behind it, stopping in the other half of the target.

The muonic x rays produced in either half of the target were detected by a single NaI crystal X, 3 in. in diameter and 3 in. thick, placed symmetrically with respect to the target. This NaI crystal was surrounded by an annular NaI crystal No. 5, 7 in. long, 3 in. inside diameter, 7 in. outside diameter. The annular counter was used in anticoincidence so as to eliminate from the x rays recorded most of those which did not lose all their energy in detector X.

The pulses from counter X were recorded and stored in one or another half (200 channels each) of the memory of a 400-channel pulse-height analyzer; a signal $123\bar{4}\bar{5}X$ directed the pulse into one half of the analyzer, and a signal $123'\bar{4}\bar{5}X$ into the other half. Long-term stability of the x-ray detection system was achieved by using a "Spectrastat" system[6] in which γ radiation from an auxiliary radioactive source (Na^{22} for the oxygen measurement, Cs^{137} for calcium, and Na^{22} for Sn and Gd) is used to correct any drift. The effectiveness of this stabilizing system is not crucial in our procedure, but its employment helped, *inter alia*, to maintain maximum energy resolution over long periods.

Spurious differences in the recorded energies of the x rays from the two isotopes could arise from inherent asymmetries: the two isotopes "see" different parts of the NaI detector, and different parts of the digital storage system of the analyzer are used to record the two spectra. To obviate these effects the NaI detector was surveyed with radioactive sources so as to determine the sensitivity variation over its surface, and it was oriented during the measurement so that the asymmetry with respect to the positions of the two isotopes was a minimum. Furthermore, the relative positions of the targets and the NaI detector were reversed in half the measurements so as to compensate for the residual asymmetry. Likewise the corresponding halves of the analyzer into which the pulses were stored were also interchanged periodically to eliminate the effects of any asymmetry from that source. All these permutations of crystal orientation, and storage section were made frequently and systematically throughout the measurements.

A further source of asymmetry in the muonic x-ray spectra might arise from the different nuclear proper-

[6] H. DeWaard, Nucleonics 13, 36 (1955).

TABLE I. Isotope target properties and experimental results.

Isotope pair	Chemical composition and isotope weight (g)	Isotopic enrichments Light	Heavy	Mean energy K_α x rays	Observed isotope shift E(light)-E(heavy) (keV)	Reduced mass correction (keV)	Corrected isotope shift (keV)
O^{16}-O^{18}	H_2O(5.0)	99.8	99.0[a]	133	-0.095 ± 0.040	-0.104	$+0.01\pm0.04$
Ca^{40}-Ca^{44}	$CaCO_3$(6.8)	96.9	98.6[b]	772	$+0.50\ \pm0.30$[c]	-0.20	$+0.70\pm0.30$
					$+0.70\ \pm0.30$[d]		$+0.90\pm0.30$
Sn^{116}-Sn^{124}	SnO_2(5.0)	92.8[b]	92.8[b]	3430	$+18\pm7$	-0.4	$+18\pm7$
Gd^{155}-Gd^{160}	Gd_2O_3(7.1)	94.3[b]	87.7[b]	4610	$+25\pm6$	-0.2	$+25\pm6$

[a] Obtained on loan from The Yeda Corporation, Rehovoth, Israel.
[b] Obtained on loan from the Isotope Development Center, Oak Ridge National Laboratory, Oak Ridge, Tennesee.
[c] This experiment.
[d] J. A. Bjorkland, S. Raboy, C. C. Trail, R. D. Ehrlich, and R. J. Powers, Phys. Rev. 136, B341 (1964).

ties of the two isotopes as displayed in the μ-capture process. Some of the nuclear gamma radiation, consequent on μ capture, could be detected by the NaI detector and this resulted in a different background for the two spectra.

Since this nuclear γ radiation is delayed (time-constant of the order of a microsecond) with respect to the μ x rays, we could examine this source of background by introducing sufficient time delay between the μ pulses $[123$ (or $3')$, $\bar{4}\bar{5}]$ and detector pulse X, so that the muonic x rays were completely suppressed and the background enhanced. We observed no differences in background of this sort which significantly affected the interpretation of the x-ray spectra.

In addition to possible asymmetries, the effectiveness of the measurements is also reduced by extraneous sources of muonic x rays which could produce an irregular background. Such x rays could arise from muons stopping in counters Nos. 3, 3′, or in the wrappings of these counters and counter No. 4 which faced the targets T and T', and in the walls of the containers used for the separated isotopes. To reduce such effects counters Nos. 3 and 3′ were $\frac{1}{16}$ in. thin, and for the wrappings we used one layer of 5-μ aluminum foil and one layer of 25-μ graphite-loaded polythene. The targets were contained in thin-walled polythene bottles or bags, identical for each pair of isotopes. Spectra were measured with empty containers instead of the usual targets. The most significant background was that due to carbon, ($K_\alpha=75$ keV, $K_\infty=100$ keV carbon) in the oxygen measurements ($K_\alpha=133$ keV), but even here the carbon background did not contribute seriously to the uncertainty of the measurements.

The chemical and isotopic composition of the material used is shown in Table I.

III. RESULTS AND ANALYSIS

The separate spectra in the region of the $2p$-$1s$ transition of each isotope, summed over the different permutations of detector orientation and analyzer arrangement, are shown in Figs. 2 through 5.

Our determination of the energy difference between the μ x rays of a pair of isotopes of the same element is based on the assumption that the two spectra are identical in all respects except for a small energy difference between the positions of the prominent peaks. All the measurements were consistent with the assumption that, when appropriately normalized, the backgrounds in the two corresponding spectra were indistinguishable. Three different procedures were used to determine the isotope shifts. In the first, the centroids of the prominent K_α line were measured after the smoothly varying background and, when possible, the contributions of K_β and higher K x rays had been subtracted by an identical procedure for each member of the pairs of isotopes. The shift is obtained from the difference in the positions of the centroids, but this "centroid" method, which is appropriate enough in the case of negligible background, is unduly sensitive to the observations far removed from the mean, and hence to background statistical fluctuations in such regions.

Since the experimental width of a single "line" in the spectrum, and in particular the $2p$-$1s$ transition, is much greater than either the width of a single channel in the analyzer or the expected energy shift between the two spectra, this shift can be determined by a simple procedure of taking differences between the spectra. The two spectra are first normalized to the same total number of counts. A region is then selected in which the number of counts varies rapidly with the channel number, i.e., the region near the peaks in the spectrum, and the differences between the two spectra in such regions, $n_1-n_2=D_n$, are recorded. From the shape of the spectral line it is easy to determine with adequate precision the rate at which n would vary if the whole spectrum is translated by a small energy change, $\delta n/\delta x$. The weighted average of $D_n/(\delta n/\delta x)$ over the appropriate regions of the spectrum provides the measure for the isotope shift. This procedure has the advantage of weighting all channels correctly but it is sensitive to any systematic difference in the shape of spectral lines. In a third method, the procedure was to subtract a given number of counts N_0 from the number in all channels and then take the centroids of the remainder. By increasing N_0 in small steps, one could then ascertain whether any apparent energy differences arose from local fluctuations or systematic differences of the two spectra. This procedure was

useful in checking whether any of the measured energy differences resulted from differences in the background in the two cases and the manner in which it was subtracted.

Although the three methods did not yield identical results the differences were small, less than the errors attributable to the limited statistical accuracy. In addition, separate analysis of individual measurements indicated no systematic errors. We have concluded therefore that our measurements are limited by random fluctuations, and it is the errors from these sources only that are listed below.

The relative significance of the several corrections and sources of error was not the same for the different elements. The specific features of the individual measurements are indicated in the following.

(*a*) *Oxygen:* O^{16}-O^{18}. (Fig. 2) In this case the expected isotope shift was the smallest of all those investigated and the precautions taken to avoid any bias were correspondingly the most stringent. The background from carbon muonic x rays as determined by measurements with empty target containers had to be subtracted carefully, and this in turn demanded a knowledge of the shape of a single line, modified by possible scattering in surrounding material. This latter was determined by recording the spectrum produced by a Co^{57} source (γ energy 122 keV) in the form of an aqueous solution in the same configuration as the water targets used (H_2O^{16} and H_2O^{18}).

(*b*) *Calcium:* Ca^{40}-Ca^{44}. (Fig. 3) The conditions here were the most favorable. The NaI detector had a sufficiently high efficiency and resolution in this energy region for the statistical uncertainties to be reduced to about the level where all systematic errors would be difficult to eliminate. The separation of the K_α and $K_{\beta\to\infty}$ x rays was clear enough for their separate contributions to be assessed and the shifts, as determined by all three methods of analysis and by using either the K_α transition alone or using the combined K transitions, were all consistent.

(*c*) *Tin:* Sn^{116}-Sn^{124}. (Fig. 4) *and Gadolinium:* Gd^{155}-Gd^{160}. (Fig. 5) For these K x rays the energies are in the region (Sn: ~3.5–4.5 MeV; Gd: ~4.7–6.3 MeV), where the response of the NaI detector to a single energy is more complex, the significant features being the full-energy peak and the "escape" peak at an energy lower by 0.51 MeV (escape of one of the annihilation rays). Our analysis of the isotope shift was based mainly on the "difference" method applied to the high-energy edge of the full-energy K_α peaks, but with some confirmation by application of the "subtraction" method to these peaks.

The final experimental results are all shown in Table I column 6.

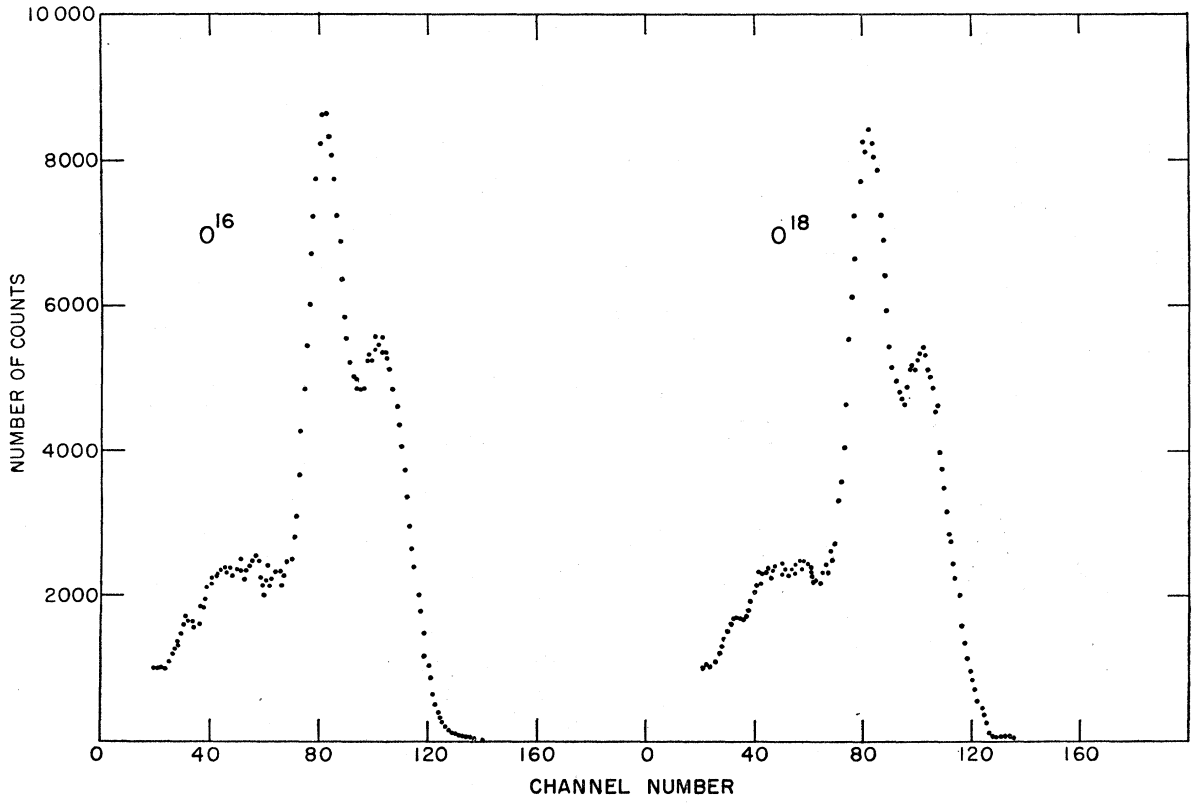

FIG. 2. Oxygen muonic x-ray spectrum after correction for background spectra.

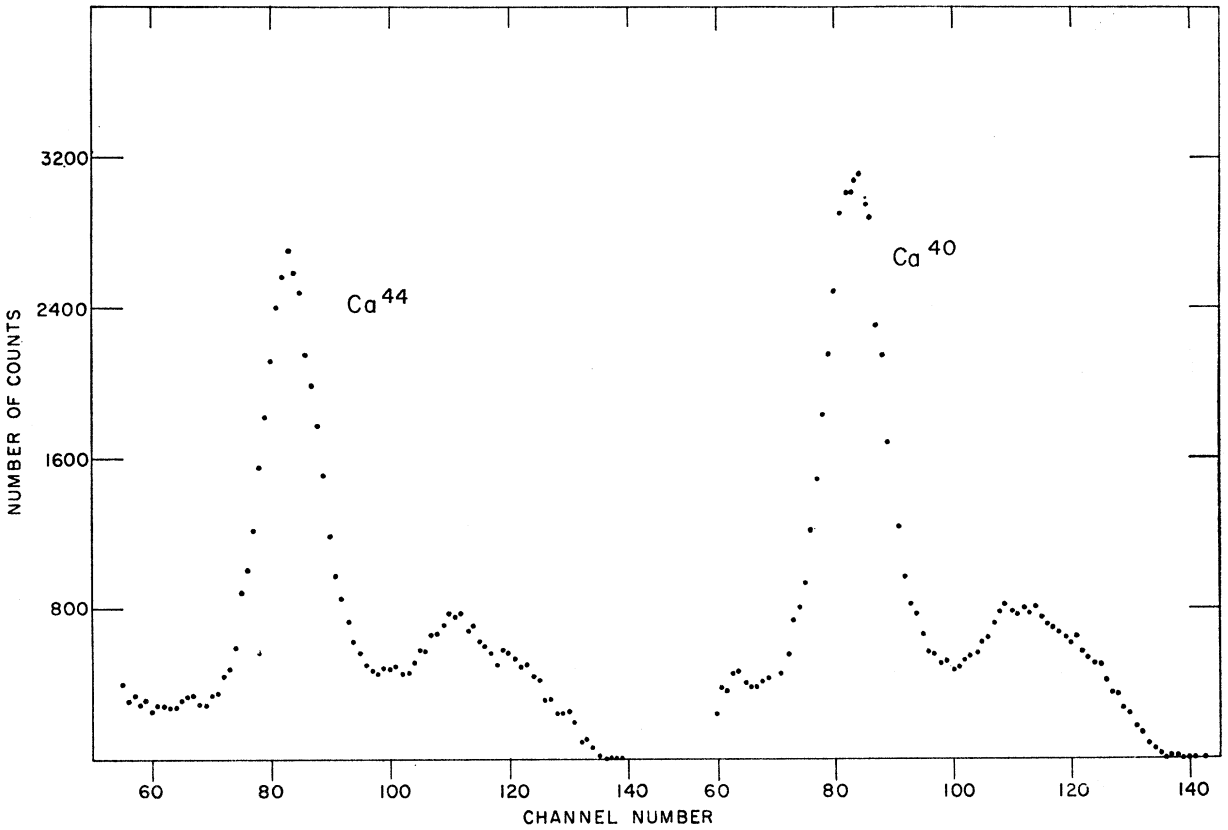

FIG. 3. Calcium muonic x-ray spectrum after correction for the background of delayed γ rays.

IV. DISCUSSION

The simplest way in which to interpret the isotope shift is in terms of the different distributions of electric charge of the two nuclei (each in the ground state). While this contribution to the isotope shift is probably the dominant effect, there are several other possibly significant contributions.

In the first place our measurements do not resolve the fine structure (and in the case of Gd^{155} the hyperfine)

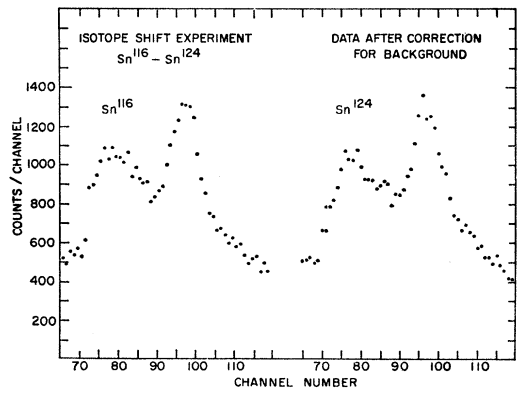

FIG. 4. Tin muonic x-ray spectrum after correction for the background of delayed γ rays.

in the spectra. The K_α x rays we observe comprise both $2p_{1/2} \rightarrow 1s_{1/2}$ and $2p_{3/2} \rightarrow 1s_{1/2}$ transitions; the relative intensities of the two constituents are determined, of course, by the relative number of $2p_{1/2}$ and $2p_{3/2}$ levels which are populated in the muonic cascade. In the case of oxygen, the fine structure is sufficiently small, in comparison with our experimental precision, to be negligible. However, for the heavier elements this is not so (Table II, column 4); but it seems difficult to see how a small change in the nuclear charge distribution could significantly affect the transition from the higher levels to the $2p$ level. It seems, therefore, reasonable to assume that any differences in the populations of the $2p$ levels are negligible in the present context: A relative change of the order 10% would be necessary to introduce errors comparable with the experimental uncertainty. With the exception of Gd^{155} ($I=\frac{3}{2}$, $Q\approx 1.6\times 10^{-24}$ cm^2), all the nuclei we have studied have zero ground-state spin and there is therefore no hyperfine splitting. In the case of Gd^{155} the estimated hyperfine displacements due to static nuclear quadrupole-muon interaction in the $2p$ level are[7]: +10 keV [$F(\equiv I+J)=1, 3$]; −30 keV ($F=2$), and +5-keV (F=0) all measured relative to the undisplaced levels, neglecting quadrupole interaction. The

[7] J. A. Wheeler, Phys. Rev. **92**, 812 (1953).

TABLE II. Calculated properties of muonic x rays and comparison with experimental results.

Isotope pair	$\frac{\Delta E/E}{\Delta R/R}$ Pustovalov formula[a]	$RA^{-1/3}\times10^{13}$ assumed in preceding column (cm)	Fine structure splitting[b] (keV)	ΔE expected for $A^{1/3}$ rule (keV)	ΔE expected from Elton's formula[c] (keV)	$\Delta E_{\text{observ}}/\Delta E_{\text{Elton}}$ for muonic x rays	$\Delta E_{\text{observ}}/\Delta E_{\text{Elton}}$ for optical isotope-shift measurements
O^{16}-O^{18}	0.022	1.35	3.8×10^{-2}	+0.10	+0.065	0.15±0.4	
Ca^{40}-Ca^{44}	0.15	1.32	1.5	+2.8	+2.1	0.24±0.15	
Sn^{116}-Sn^{120}	0.50	1.20	46	+39	+34	0.53±0.18	0.50±0.20[d]
Gd^{155}-Gd^{160}	0.63	1.20	96	+32	+29	0.86±0.20	0.94±0.20[e]

[a] Reference 20 (1959).
[b] K. W. Ford and J. G. Wills, Nucl. Phys. **35**, 295 (1962); Los Alamos Laboratory Report, LAMS-2387, 1962 (unpublished).
[c] Reference 22.
[d] D. N. Stacey, Proc. Roy. Soc. (London) **A280**, 439 (1964).
[e] H. Kopfermann, Fruger, and Steudel, Ann. Physik **20**, 258 (1957).

centroid of the $2p$ level is not displaced by this quadrupole interaction and since the displacements are small relative to the resolution of our detecting equipment (~400 keV for Gd μ x rays) we can still treat the complex transition as a single one.

The estimated magnetic hyperfine interaction is very much smaller than the electric-quadrupole. In treating the isotope shifts as due to different static-charge distributions, one is ignoring the dynamic properties of the nucleus, i.e., the influence of the muon on internal degrees of freedom of the nucleus. Since we are only attempting to interpret the isotope shift, and not the absolute muonic x-ray energies, we are only concerned with the *difference* in this interaction as between one isotope and another. There have been numerous attempts to estimate these so-called "nuclear polarization" effects, using widely different nuclear models and different computational approximations. For those models in which the polarization, in common with other nuclear properties, is treated as a smoothly varying function of A and Z, then the change in polarizability arising from the small difference in A is usually very small: e.g., in Cooper and Henley's calculations,[8] using an independent-particle model which apparently overestimates the effect, the contribution to the isotope shift is 6 eV for O^{16}-O^{18}, and 200 eV for Ca^{40}-Ca^{44}, and insignificant amounts for Sn and Gd.

Nuclear polarization effects have also been examined using nuclear models, such as the hydrodynamical model, which are the opposite extreme as far as nucleon correlations are concerned, but which retain the feature that polarizability is a smoothly varying, "average" nuclear property (e.g., Steinwedel and Jensen,[9] Lakin and Kohn,[10] Nuding,[11] Greiner[12] and Scheck[13]). The general conclusion from these investigations appears to be that, on account of correlation of nucleon motion in the nucleus (as exemplified in the hydrodynamical model), the contribution of nuclear polarization is overestimated in the calculation of Cooper and Henley. The tentative conclusion is then that the difference in charge distribution is the predominant feature of the isotope shift, and that it is pertinent therefore to compare the experimental results with this feature.[14–17]

In all cases a straightforward connection has to be made for the finite nuclear mass. This reduced-mass correction is shown in Table I, column 7.

In principle, one should start with an assumed nuclear charge distribution and compute the corresponding energy levels of the muonic atom. Practically, these energy levels, in the lighter nuclei at least,

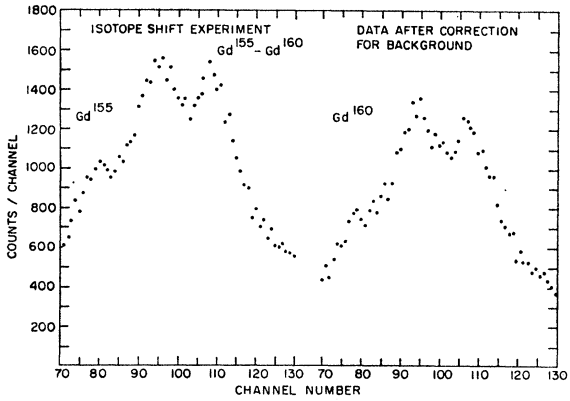

FIG. 5. Gadolinium muonic x-ray spectrum after correction for the background of delayed γ rays.

[8] L. Cooper and E. Henley, Phys. Rev. **92**, 801 (1953); D. L. Hill and K. W. Ford, *ibid.* **94**, 1617 (1954).
[9] V. H. Steinwedel and J. H. D. Jensen, Z. Naturforsch. **5a**, 413 (1950).
[10] W. Lakin and W. Kohn, Phys. Rev. **94**, 787 (1954).
[11] E. Nuding, Z. Naturforsch. **12a**, 187 (1957).
[12] W. Greiner, Z. Physik **164**, 374 (1961).
[13] F. Scheck, Z. Physik **172**, 239 (1963).

[14] Substantial energy differences could arise, of course, if the nucleus were left excited by the muonic transitions, a possibility for deformed nuclei with large intrinsic quadrupole moments and low-lying excited states. Estimates of this effect have been made by Wilets (Ref. 15) and Jacobson (Ref. 16) and the effect has also been observed (Ref. 17). However, for the nuclei with which we are concerned the differences in energy between the ground and the lowest excited states are quite large, compared to the $2p_{3/2}$-$2p_{1/2}$ splitting, so that nuclear excitation is not expected to be appreciable.
[15] L. Wilets, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **29**, No. 3 (1954).
[16] B. A. Jacobson, Phys. Rev. **96**, 1637 (1954).
[17] R. D. Ehrlich, R. J. Powers, V. L. Telegdi, J. A. Bjorkland, S. Raboy, and C. C. Trail, Phys. Rev. Letters **13**, 550 (1964).

depend only on the mean square radius of the charge distribution: $R^2=\langle r^2\rangle$. Any reasonable estimate of the change in the shape of the radial charge distribution from one isotope to another indicates that for the limited precision and resolution of our measurements, it would not be meaningful to consider such finer details of the distribution. The crudest basis for comparison would be to take the charge distribution as uniform throughout the nuclear volume and assume the latter to be spherical with a radius proportional to $A^{1/3}$. There is, of course, no experimental basis for applying such a simple dependence to the variation of charge radius of different isotopes of the same element. It is interesting to note that the relative size of the nuclei Ca^{40} and Ca^{44}, as determined by the diffraction of 30-MeV α particles, does seem to be consistent with an $A^{1/3}$ dependence.[18]

It is well known from optical isotope-shift measurements of atomic spectra that the successive addition of neutrons to a nucleus does not result in a uniform increase in the nuclear radius. Especially well established is the "odd-even" effect: The addition of the first, third, etc., neutron to an even nucleus produces a much smaller expansion than that of the second, fourth, etc. Each successive *pair* of neutrons does appear to result in a similar degree of expansion; but as mentioned in the Introduction an absolute, quantitative estimate of this change in size requires reliable knowledge of atomic wave functions. In the present experiments the wave functions are essentially known; the limitation is in the precision with which the isotope shift has been measured.

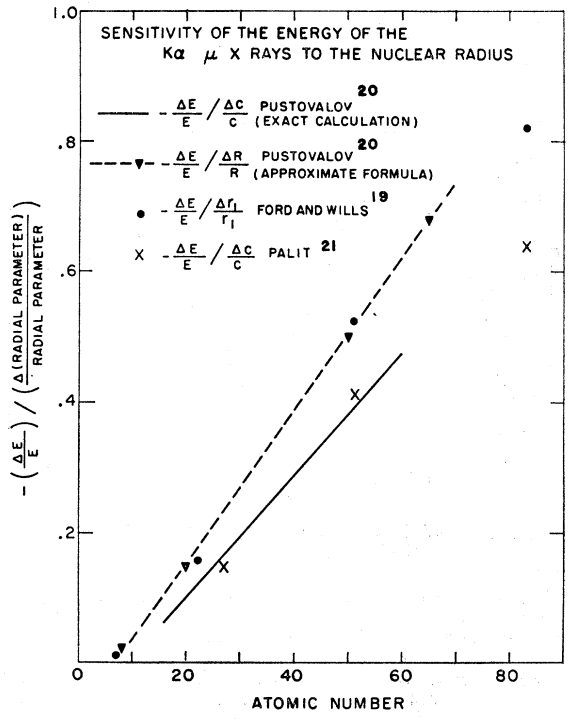

FIG. 6. Sensitivity of the energy of the K_α muonic x rays to variations of the nuclear radius: $(-\Delta E/E)/(\Delta R/R)$ as a function of the atomic number according to the calculation of Pustavalov, Palit, and Ford and Wills.

[18] N. S. Wall, G. Heyman, R. W. Bauer, and A. M. Bernstein (private communication); reported at the Paris Conference on Nuclear Structure, June 1964 (unpublished).

To interpret the measured isotope shifts in terms of changes in nuclear radii (subject to the uncertainties already discussed) we need to assume a mean nuclear radius for each isotope pair. Extensive calculations of the muonic K_α x-ray energies as a function of Z and nuclear radius have been made by Ford and Wills,[19] Pustovalov,[20] and Palit.[21] A rough indication of the sensitivity of the isotopic shift to changes in nuclear radius is shown if Fig. 6 and in Table II, column 5 which is based on Pustovalov's calculations and a nuclear radius $R=r_0A^{1/3}\times10^{-13}$ cm, with r_0 taken from electron scattering measurements.

More elaborate empirical expressions for the variation of nuclear radius with A, still based on nuclei of maximum stability, have also been proposed. In particular, Elton[22] has found the formula

$$R(A)=1.115A^{1/3}+2.151A^{-1/3}-1.742A^{-1}$$

gives a good empirical fit to observations—chiefly electron scattering—when the nuclear density distribution is taken to be

$$\rho(r)=\rho(0)[1-\exp(-c/d)]/(1+\exp[(r-c)/d]).$$

The central density $\rho(0)$ and the surface thickness d are both assumed independent of A, and neutron and proton distributions are identical, i.e., no distinction is made between nuclear radius and nuclear charge radius. This formula for $R(A)$ "predicts" a rather smaller isotope shift than the $A^{1/3}$ rule (Table II, column 6). Although this empirical formula has no physical justification when applied to a variation of neutron number alone, it is convenient to express both the muonic isotope shift and the optical isotope shifts in terms of those predicted by it. In two cases, Sn^{116}-Sn^{124} and Gd^{155}-Gd^{160}, a direct comparison can be made between the muonic and optical-atomic measurements (Table II, column 8).

In the case of tin both measurements indicate that $\Delta R/R$ is up about one-half the value "predicted" by the empirical $R(A)$ formula. For gadolinium, both measurements are consistent with a shift of 0.9 of the empirical "predictions."

[19] K. W. Ford and J. G. Wills, Phys. Rev. **94**, 1680 (1954).
[20] G. E. Pustovalov, Zh. Eksperim. i Teor. Fiz. **43**, 2170 (1962); **36**, 1806 (1959) [English transl.: Soviet Phys.—JETP **16**, 1534 (1963); **9**, 1288 (1959)].
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The fact that the change in nuclear radii caused by addition of pairs of neutrons (the odd-even effect is attributable to other details of nuclear structure), is less than the empirical formulas predict—either the crude $A^{1/3}$ or the more elaborate expression—has been examined in terms of the possible effects of nuclear compressibility and changes in surface and volume-symmetry energies. Bodmer's[23] detailed examination of this matter suggests a much smaller compressibility energy than might be expected for general considerations. Additional evidence for such a "soft" nucleus is also provided by the analysis (Greiner and Scheck[24]) of muonic x-ray measurements of neighboring elements by Quitman *et al.*[25]

We conclude that the muonic isotope shifts confirm some of the conclusions of the optical measurements, and in principle enable this type of investigation to be extended to lighter nuclei. The development of improved γ-detection techniques now makes it possible to consider more extensive and more precise measurements.

Note added in proof. Recent measurements of the difference in the scattering of 250-MeV electrons by Ca^{40} and Ca^{44} indicate a difference in charge distribution for the two nuclei which is in good agreement with that reported in this paper. [R. Hofstadter *et al.*, Phys. Rev. Letters **15**, 758 (1965).]

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Hyperfine Separation of Deuterium*

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The hyperfine separation of deuterium has been measured by a spin-exchange technique in which deuterium interacts with radiating hydrogen in a hydrogen maser. Resonance of the deuterium is detected by its effect on the hydrogen oscillation power level. The result is $\Delta\nu(\mathrm{D}) = 327\,384\,352.3 \pm 0.25$ cps in the $A1$ time scale [$\Delta\nu(\mathrm{Cs}) = 9\,192\,631\,770$ cps]. A theoretical analysis of the technique and experimental details are presented.

I. INTRODUCTION

THIS paper describes a redetermination of the ground-state hyperfine separation of deuterium by a new technique in which a hydrogen maser[1–3] is used as a polarization detector. The maser oscillates on the hydrogen hyperfine transition $(F=1, m_F=1) \rightarrow (F=0, m_F=0)$ at approximately 1420 Mc/sec. Spin-exchange collisions between deuterium and hydrogen relax the oscillating hydrogen magnetic moment by an amount depending on the deuterium electron polarization as well as on the spin-exchange collision rate. When a radiation field is applied at a deuterium resonance frequency, the deuterium electron polarization decreases and there is a corresponding decrease in the oscillating hydrogen magnetic moment. Changes in the maser power level thus serve to detect deuterium resonances.

This technique is similar to the optical-pumping spin-exchange method[4] in that changes in the deuterium electron polarization are detected by spin-exchange coupling to a second spin system whose electron polarization is more conveniently observed. However, it preserves two advantages of the hydrogen maser: The frequency shifts due to wall collisions are small relative to those produced by buffer-gas collisions in an optical-pumping experiment, and better spatial averaging of the magnetic-field gradients reduces the resonance linewidth and produces a highly symmetric line.

In principle, the deuterium hyperfine separation could have been measured with equal precision using a deuterium maser. However, there are substantial experimental difficulties associated with construction of such a maser owing mainly to the relatively low deuterium hyperfine frequency. The present method was chosen for its experimental simplicity, its interesting

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