

ISOTOPE SHIFTS IN MUONIC X RAYS OF  $\text{Sn}^{118, 119, 120}$ ,  $\text{Nd}^{142, 144, 146}$ , AND  $\text{W}^{182, 184, 186}$ †T. T. Bardin, R. C. Barrett,\* R. C. Cohen, S. Devons, D. Hitlin, E. Macagno,  
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(Received 11 March 1966)

One of the effects of the extended charge distribution in the nucleus is to raise the positions of bound electron levels in an atom by an amount  $\Delta E$  from those due to a point-charge nucleus. The small shifts from isotope to isotope,  $\delta\Delta E$ , in the energy levels of an atom result in the well-known and extensively studied phenomenon of optical "isotope shifts." Similar effects are expected, of course, in muonic x-ray spectra.<sup>1</sup> In the latter case, however, the effects are very much larger on account of the relatively larger muon probability amplitude within the nucleus; therefore, muonic x-ray transition energies are most sensitive to the finite extension of the nuclear charge. Furthermore, the muonic energy levels are not complicated by screening effects and can be accurately calculated for any given charge distribution. As part of the study of the relation between these two sets of isotope shift (optical and muonic), we have studied the muonic x-ray spectra of the following group of isotopes:  $\text{Sn}^{118, 119, 120}$ ,  $\text{Nd}^{142, 144, 146}$ , and  $\text{W}^{182, 184, 186}$ , all of which have been studied optically.<sup>2</sup> The imprecise knowledge of the many-electron atomic wave functions, including the problem of completely separating the kinematical mass effects from the nuclear field effects, restrict the full analysis of optical isotope shifts to the measurement of the ratios of the  $\delta\Delta E$  values within a sequence of isotopes. Absolute values of energy levels are not derived from the observations, nor can the interpretation of the differences between particular isotopes exploit the full experimental precision of the relative measurements. By contrast, isotope shifts in muonic x-ray spectra, which can now be measured with a precision approaching that of optical shifts (provided sufficient quantities of separated isotopes are available), are capable of more specific interpretation in terms of nuclear-field parameters. For these reasons, then, the two types of measurements—optical and muonic—are largely complementary: The precise, but relative, optical measurements, which can be made even with minute quantities<sup>3</sup> of isotope (as few as  $10^{12}$  atoms) can be normalized and

more completely interpreted when supplemented by some "absolute" muonic measurements of the same elements.

The elements we have investigated are known to display markedly different features optically, and the muonic x-ray properties would be expected to show differences at least as pronounced.

The major trends of the optical isotope shift results are shown in the well-known Brix-Kopferman diagram<sup>4</sup> (Fig. 1), which exhibits the measured effect of adding pairs of neutrons, in different atoms. The shift is expressed as a ratio of the observed values  $\delta(\Delta E)_{\text{exp}}$  to those calculated on the basis of a spherical nucleus with uniform charge density and radius proportional to  $A^{1/3}$ ,  $\delta(\Delta E)_{\text{std}}$ . The calculated value  $\delta(\Delta E)_{\text{std}}$  from the over-simplified one-parameter nuclear model was originally used only as a convenient reference magnitude to normalize the observed shifts in optical results.

The main feature of the optical isotope shifts can be summarized<sup>4,5</sup> as follows: (a) The majority of optical isotope shifts  $\delta(\Delta E)_{\text{exp}}$  are about  $\frac{1}{2}$  or less than  $\delta(\Delta E)_{\text{std}}$ . (b) The fluctuations

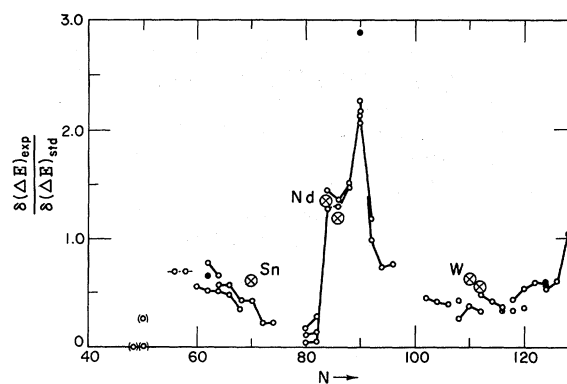


FIG. 1. Brix-Kopferman diagram with muonic x-ray experimental results ( $\otimes$ ) superimposed on it.  $\delta(\Delta E)_{\text{exp}}/\delta(\Delta E)_{\text{std}}$  is the observed isotope shift expressed in terms of a so-called standard isotope shift which is the shift calculated on a spherical nucleus model of uniform charge distribution with radius  $R = 1.19 \times 10^{-13} A^{1/3}$ . Points are plotted against neutron number  $N$  and are placed at the higher  $N$  value for each pair of isotopes,  $\text{Sn}^{118-120}$ ,  $\text{Nd}^{142-144}$ ,  $\text{Nd}^{144-146}$ ,  $\text{W}^{182-184}$ , and  $\text{W}^{184-186}$ .

of the ratio  $\delta(\Delta E)_{\text{exp}}/\delta(\Delta E)_{\text{std}}$  are closely correlated with nuclear magic numbers. The theoretical interpretation<sup>4-6</sup> of these trends is that they result from the combination of two factors: the change in the nuclear volume on adding neutrons, which therefore reduces the actual charge density of the nucleus, and the change in nuclear deformation, which, averaged over all angles, has the effect of extending the equivalent radius, i.e., the radius of a uniform charge distribution which leads to the same transition energy. These two effects can be in the same or opposite senses. For Nd, these two effects are in the same sense, as the lightest isotope  ${}_{60}\text{Nd}_{82}^{142}$  has a closed neutron shell, therefore the deformation increases as more neutrons are added. However, these effects are in the opposite sense in W isotopes as the nuclear distortion decreases as more neutrons are added.

The results of our muonic x-ray measurements, of which details are given below, are marked by  $\otimes$  in Fig. 1, and it is clear that they follow the trends expected from the optical work. An additional feature of the optical results is the odd-even staggering: The levels of an odd-mass isotope lie much closer to its lighter than to its heavier even-even isotope. This feature is also exemplified in the muonic measurements of the Sn isotopes.

The experimental arrangements used are similar to those described in an earlier Letter.<sup>7</sup> The accuracy of the measurements is limited partly by statistical fluctuations, and partly by systematic effects (drifts in sensitivity, etc.). The latter could, in principle, have been eliminated by using the system of simultaneous observations of pairs of isotopes with identification by electronic logic which has been described in a previous communication<sup>8</sup>; but in the present context, with small detectors and limited available target material (separated isotopes),

Table I. Muonic x-ray target compositions.

Elements	Weight (g)	Surface		Chemical composition
		density (g/cm <sup>2</sup> )	Enrichment (%)	
Sn <sup>118</sup>	45.604	2.25	97.1	Metallic powder
Sn <sup>119</sup>	28.959	1.43	89.0	
Sn <sup>120</sup>	149.876	7.39	98.3	Nd <sub>2</sub> O <sub>3</sub> packed in sulphur
Nd <sup>142</sup>	182.648	1.08	96.03	
Nd <sup>144</sup>	180.480	1.07	94.5	Metallic powder
Nd <sup>146</sup>	131.707	0.78	94.5	
W <sup>182</sup>	80.160	3.95	94.7	
W <sup>184</sup>	59.160	2.92	94.3	
W <sup>186</sup>	44.407	2.19	97.2	

the less favorable geometry and the consequent loss of statistical accuracy would have outweighed the improvement. To reduce the systematic errors to a minimum, targets were interchanged frequently and the measurements were interspersed with frequent calibration, so that the effects of drifts and fluctuations were estimated to be the order of 1 keV or less. The accuracy of the over-all measurements is limited chiefly by statistical fluctuations.

The isotopic enrichment, the surface density, and the chemical composition of the targets used are listed in Table I.

Sn<sup>118, 119, 120</sup>.—The measurements of the  $2p_{3/2} \rightarrow 1s_{1/2}$  and  $2p_{1/2} \rightarrow 1s_{1/2}$  transitions for the three tin isotopes are summarized in Table II. In Table III, the observed isotope shifts  $\delta(\Delta E)_{\text{exp}}$  are displayed together with the calculated volume effect which is the so-called standard isotope shift  $\delta(\Delta E)_{\text{std}}$  based on a spherical nucleus of uniform charge distribution with radius  $R = 1.19 \times 10^{-13} A^{1/3}$ .<sup>9</sup> When the observed isotope shift between Sn<sup>118</sup> and Sn<sup>120</sup> is compared to the calculated volume effect, the ratio is only around 0.6. Also, the odd-even staggering is very pronounced here.

Table II. The experimental muonic K and L x-ray energies in keV, in Sn<sup>118, 119, 120</sup> and Nd<sup>142, 144, 146</sup>.

Z	Element	K x rays		L x rays		
		$2p_{3/2} \rightarrow 1s_{1/2}$	$2p_{1/2} \rightarrow 1s_{1/2}$	$3d_{3/2} \rightarrow 2p_{1/2}$	$3d_{5/2} \rightarrow 2p_{3/2}$	$3d_{3/2} \rightarrow 2p_{3/2}$
50	Sn <sup>118</sup>	3459.5 ± 1	3414 ± 1			
	Sn <sup>119</sup>	3459 ± 1	3415 ± 1			
	Sn <sup>120</sup>	3454 ± 1	3408 ± 1			
60	Nd <sup>142</sup>	4356 ± 1	4275 ± 1	1472 ± 1	1404 ± 1	1389 ± 1.5
	Nd <sup>144</sup>	4338 ± 1	4258 ± 1	1472 ± 1.5	1402 ± 1.5	
	Nd <sup>146</sup>	4324 ± 1	4242 ± 1	1470 ± 1.5	1402 ± 2.0	

Table III. Tabulation of the observed isotope shifts  $\delta(\Delta E)_{\text{exp}}$ , the so-called standard isotope shifts  $\delta(\Delta E)_{\text{std}}$ , and their ratios  $\delta(\Delta E)_{\text{exp}}/\delta(\Delta E)_{\text{std}}$  in the isotope pairs  $\text{Sn}^{118-119}$ ,  $\text{Sn}^{118-120}$ ,  $\text{Nd}^{142-144}$ ,  $\text{Nd}^{144-146}$ ,  $\text{W}^{182-184}$ , and  $\text{W}^{184-186}$ .

Pairs of isotopes	$\delta(\Delta E)_{\text{exp}}$ (keV)		$\delta(\Delta E)_{\text{std}}$ (keV)		$\delta(\Delta E)_{\text{exp}}/\delta(\Delta E)_{\text{std}}$	
	$2p_{1/2} \rightarrow 1s_{1/2}$	$2p_{3/2} \rightarrow 1s_{1/2}$	$2p_{1/2} \rightarrow 1s_{1/2}$	$2p_{3/2} \rightarrow 1s_{1/2}$	$2p_{1/2} \rightarrow 1s_{1/2}$	$2p_{3/2} \rightarrow 1s_{1/2}$
$\text{Sn}^{118}-\text{Sn}^{119}$	$-1 \pm 2$	$+0.5 \pm 2$	4.9	4.9	0.2	0.1
$\text{Sn}^{118}-\text{Sn}^{120}$	$6 \pm 2$	$5.5 \pm 2$	9.9	10.0	0.6	0.6
$\text{Nd}^{142}-\text{Nd}^{144}$	$17 \pm 2$	$18 \pm 2$	12.4	12.7	1.3	1.4
$\text{Nd}^{144}-\text{Nd}^{146}$	$16 \pm 2$	$14 \pm 2$	12.3	12.6	1.3	1.1
$\text{W}^{182}-\text{W}^{184}$	$10 \pm 2$	$9 \pm 2$	14.5	15.1	0.7	0.6
$\text{W}^{184}-\text{W}^{186}$	$9 \pm 2$	$7 \pm 2$	14.4	15.0	0.6	0.5

$\text{Nd}^{142, 144, 146}$ .—In Fig. 2, the  $K$  x rays of the three Nd isotopes are shown one above the other. The isotope shifts are markedly displayed. Both  $K$  and  $L$  muonic x-ray measurements are summarized in Table II. It is clear that the isotope shifts of the  $2p$  states are too small to be measured reliably. The shifts are shown in Table III, and the ratios, calculated as in the case of Sn, exhibit anomalously large values which are comparable to the optical shifts.

$\text{W}^{182, 184, 186}$ .—The isotope shifts in W isotopes are more involved because not only are the  $1s$  levels perturbed by the finite extent of the nucleus, but also the  $2p$  levels are shifted due to the dynamic  $E2$  interactions. The dynamic  $E2$  effects in highly deformed muonic atoms have been investigated theoretically by Jacobsohn<sup>10</sup> and independently by Wilets.<sup>11</sup> This effect in W isotopes has been experimentally studied<sup>12</sup> in our laboratory. The perturbation on the  $2p$  energy levels can be easily deduced from the measurements. For example, the measured isotope shift of the hfs line No. 1,  $2p_{1/2}(I=0) - 1s_{1/2}(I=0)$ , between  $\text{W}^{182}$  and  $\text{W}^{184}$  is 5.8 keV. The calculated difference of energy shifts of the level  $2p_{1/2}(I=0)$  in  $\text{W}^{182, 184}$  is  $21.6 - 17.3 = 4.3$  keV. Therefore, correcting for the difference in the  $2p_{1/2}$  hyperfine shifts yields  $\delta\Delta E_{\text{exp}} = 5.8 + 4.3 = 10.1$  keV. The results for W isotopes are summarized also in Table III.

The precise measurements of  $2p - 1s$  transitions by the NaI detectors in earlier experiments gave only a single accurate parameter which may be called the equivalent radius. Since the fine structure of both  $K$  and  $L$  x rays can now be resolved and determined by Ge(Li) detectors, this additional information should yield more than one parameter for describing the detailed charge distribution. For example, if we assume a Fermi-type charge distribution of the

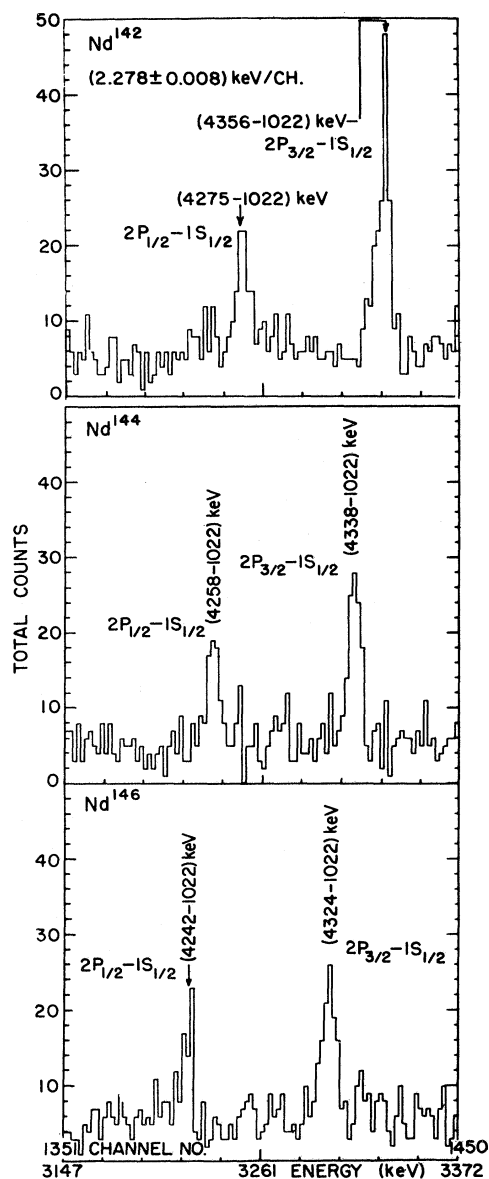


FIG. 2.  $K$  x-ray spectra of  $\text{Nd}^{142}$ ,  $\text{Nd}^{144}$ , and  $\text{Nd}^{146}$ .

nucleus and arbitrarily choose a radius of  $1.10 \times A^{1/3}$  F for the half-density radius  $C$  of the charge distribution, then the  $2p-1s$  x-ray energies in Nd give us the following values of the diffuseness parameter:  $z = 0.558 \pm 0.002$ ,  $0.568 \pm 0.002$ , and  $0.576 \pm 0.002$  for  $A = 142$ ,  $144$ , and  $146$ , respectively. These values are based on the assumption of a normal volume effect with Fermi-type charge distribution. The actual increases in the apparent diffuseness are likely to be considerably larger, since the observed experimental effect is usually smaller than the "standard" volume effect; in the average the ratio is  $\frac{1}{2}$ . The  $2p-1s$  x-ray energies for W give values of the diffuseness parameter  $z = 0.590 \pm 0.003$ ,  $0.587 \pm 0.003$ , and  $0.581 \pm 0.003$  for  $A = 182$ ,  $184$ , and  $186$ , respectively. For  $\text{Nd}^{142}$ , we can obtain a second parameter in the charge distribution from the  $3d-2p$  transition energies. The limits on the radius and the diffuseness thus obtained are  $(1.131 \pm 0.011)A^{1/3}$  F and  $0.467 \pm 0.035$  F. For the other isotopes the corresponding parameters can be determined only within wide limits.

An improvement in accuracy by a factor of 5 is quite feasible in future studies of muonic x rays. This investigation already shows the great potential of muonic x-ray studies in the determination and interpretation of isotope shifts.

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†Work partially supported by the U. S. Atomic Energy Commission and the U. S. Office of Naval Research.

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