PHYSICAL REVIEW C VOLUME 17, NUMBER 5 MAY 1978

Variations of nuclear charge radii in mercury isotopes with $A = 198, 199, 200, 201, 202$, and 204 from x-ray isotope shifts

P. L. Lee
Department of Physics and Astronomy, California State University, Northridge, California 91330
and California Institute of Technology, Pasadena, California 91125

F. Boehm and A. A. Hahn
California Institute of Technology, Pasadena, California 91125

(Received 18 October 1977)

The isotope shifts of atomic K x rays were measured for pairs of the six mercury isotopes with $A = 198$, 199, 200, 201, 202, and 204, using a curved crystal spectrometer. The changes of the nuclear charge radii were derived in terms of $\delta\langle r^2\rangle$ and δR_k and compared with optical and muonic isotope shift data. From our results, a renormalization of the optical data was obtained.

[NUCLEAR STRUCTURE x rays, ^{198,199,200,201,202,204}Hg; measure K x-ray isotope shifts enriched targets; derived $\delta\langle r^2\rangle, \delta R_k$.]

I. INTRODUCTION

Detailed investigations of the isotope shifts of optical transitions in mercury, with its large number of isotopes, have been carried out in recent years. The results of these measurements up to 1973 have been summarized by Heilig and Steudel.[1] More recent experiments on the neutron deficient Hg isotopes have been performed by Otten *et al.*[2] using the optical pumping method. This latter work has given evidence of a dramatic increase in the magnitude of the isotope shift (IS) for several low neutron number isotopes and has triggered theoretical discussions of sizable nuclear deformations in mercury.

The analysis of the optical IS data in terms of nuclear parameters is complicated by the uncertainties in the electronic wave function at the nucleus. These wave functions, calculated by atomic Hartree-Fock methods, suffer from a considerable ($\leq 10\%$ in Hg)[1] uncertainty. Specific mass shift corrections have not been calculated but from the close agreement in the relative IS it can be concluded that these corrections are small.

The optical data can be converted to a more reliable absolute scale with the help of the K x-ray IS's which are free of uncertainties from atomic calculations. We have therefore investigated the K x-ray IS covering the six stable isotopes 198, 199, 200, 201, 202 and 204 of mercury. The K x-ray IS between ^{200}Hg and ^{204}Hg has been reported previously.[3]

The energy shift of K x-ray transitions between two isotopes A and A' in medium-heavy and heavy elements is a direct measure of the change in the nuclear monopole Coulomb field. This Coulomb shift $\delta E^{A,A'}_{\text{Coul}}$, can be related[4] to the variations in the even charge moments $\delta\langle r^{2N}\rangle$ of the nucleus by defining a parameter λ,

$$\lambda \equiv \frac{\delta E^{A,A'}_{\text{Coul}}}{C_1} = \delta\langle r^2\rangle + \frac{C_2}{C_1}\delta\langle r^4\rangle + \frac{C_3}{C_1}\delta\langle r^6\rangle + \ldots$$
$$\approx \delta\langle r^2\rangle, \qquad (1)$$

where the C_i's are electronic coefficients given in Ref. 4. A small contribution arising from the dynamics of the electron motion, (mass shift), can be easily calculated. For details we refer the reader to a review of the subject in Ref. 5.

II. EXPERIMENT AND RESULT

Measurements of the differences of the $K\alpha_1$ x-ray energies of six Hg isotopes were performed with the Caltech 2-m radius bent-crystal spectrometer in the Cauchois geometry. The experimental setup and data analysis have been described in Refs. 3 and 6.

The isotope samples were obtained on loan from Oak Ridge National Laboratory. The isotopic com-

TABLE I. Percentage composition of the six Hg samples.

Sample \ Isotopes	Isotopic composition (%) 198	199	200	201	202	204
198	71.7	22.0	3.9	1.0	1.2	0.2
199	1.6	91.5	5.0	0.8	1.1	0.2
200	2.5	6.1	75.4	5.7	9.0	1.2
201	0.2	0.4	2.2	92.3	4.8	0.2
202	0.1	0.2	0.5	1.4	97.6	0.3
204	3.7	6.1	8.3	5.0	12.8	64.2

TABLE II. Isotope shifts $\delta E = E(A) - E(A')$ for Hg (in meV), upper entries, and change of the nuclear charge radius $\lambda \approx \langle r^2\rangle_A - \langle r^2\rangle_{A'}$, [cf. Eq. (1)] in fm^2, lower entries.

A' \ A	199	200	201	202	204
198	−42(19) 0.027(12)	−162(41) 0.103(26)	−182(26) 0.116(17)	−312(39) 0.199(25)	−468(44) 0.298(28)
199		−120(36) 0.076(23)	−140(20) 0.089(13)	−270(35) 0.172(22)	−425(40 0.271(26)
200			−20(32) 0.013(20)	−150(12) 0.095(8)	−305(30) 0.194(19)
201				−130(29) 0.082(18)	−286(36) 0.182(23)
202					−156(44) 0.099(28)

position of the six samples is listed in Table I. Chemical and crystallographical purity of the samples were checked with a Guinier camera.

According to the criterion given in Ref. 3, an optimum amount of enriched HgO powder was mixed with Formvar powder and pressed into a pill. Several series of measurements were performed using combinations of three or four isotopes in each series.

We refer to Refs. 3 and 6 for the details concerning the line fitting procedures and data analysis. As shown in Table I, the isotope samples for ^{198}Hg and ^{200}Hg and ^{204}Hg contain appreciable amounts of the other Hg isotopes. Accordingly, a significant correction had to be applied to the observed shift. Since we had measured all the isotopes involved, this can be done exactly by writing each sample shift as a weighted (according to isotopic composition) average of the true IS. The IS was recovered by inverting these equations. The final IS values were obtained by a least-squares fit to all the measured combinations. A small correction for the mass shift (1.3 meV per two neutrons)[3] was applied to the data. The final results for the IS are presented in Table II. The uncertainties quoted represented one standard deviation of the experimental uncertainties. The charge radius variation $\lambda \approx \delta\langle r^2\rangle$ calculated from Eq. (1) using $C_1 = 1.57 \times 10^3$ meV/fm^2 is also shown in Table II.

III. COMPARISON WITH OPTICAL AND MUONIC ISOTOPE SHIFTS

The present results can be compared with optical IS for all the Hg isotopes[1,2] as well as muonic IS of the six stable isotopes from ^{198}Hg to ^{204}Hg,

TABLE III. $\delta R_k = R_k(A) - R_k(A')$ for atomic K x rays [Equation (2) with $k = 2.917$, $C_Z = -78.1 \times 10^{-6}$ fm/meV] (top entry), optical transitions [$\delta\nu = -44.9(5.0)(\mathrm{GHz/fm^2})\delta\langle r^2\rangle$ (Refs. 1 and 2)] (middle entry), and muonic x rays [Equation (2) with $k = 2.413$, $C_Z = -1.328 \times 10^{-3}$ fm/keV (Ref. 8)] (bottom entry) for each pair of isotopes. The units are 10^{-3} fm.

A' \ A	199	200	201	202	204
198	3.3(1.5) 1.8(2) 1.97(29)	12.7(3.2) 13.2(1.5) 11.55(27)	14.2(2.0) 17.7(2.0) 16.53(47)	24.4(3.0) 27.7(3.1) 24.51(28)	36.6(3.5) 42.1(4.7) 36.75(37)
199		9.4(2.8) 11(1) 9.59(32)	10.9(1.6) 15.8(1.7) 14.57(49)	21.1(2.7) 25.9(2.8) 22.55(31)	33.2(3.1) 40.2(4.5) 34.78(37)
200			1.5(2.5) 4.4(5) 4.98(46)	11.7(1.0) 14.5(1.6) 12.96(25)	23.8(2.3) 28.8(3.2) 25.19(33)
201				10.2(2.3) 10.1(1) 7.98(45)	22.3(2.8) 24.4(2.7) 20.21(49)
202					12.2(3.4) 14.3(1.6) 12.23(27)

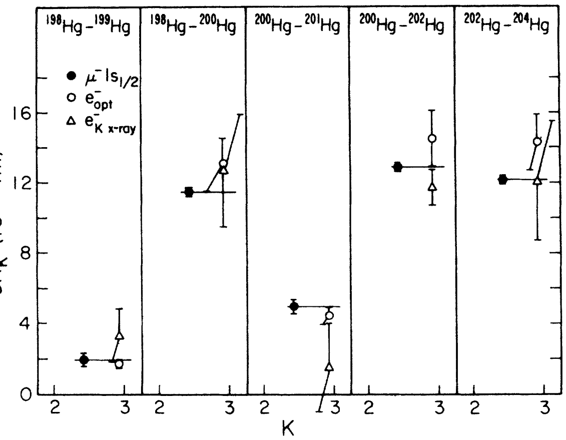

FIG. 1. Comparison of μ^- x ray, e^- x ray, and optical IS. The solid line is calculated from muonic data (Ref. 7) assuming a Fermi distribution for the nuclear monopole charge density. The e^-_{opt} data have been calculated using the conversion factor $\delta\nu = -44.9(5.0)$ GHz/fm^2 (Ref. 1).

recently measured by Hahn *et al.*[7,8] by resorting to the equivalent radius R_k, defined[9] by the model independent moment

$$\langle e^{-\alpha r} r^k \rangle = \frac{3}{R_k^3} \int_0^{R_k} e^{-\alpha r} r^{k+2}\, dr$$

$$= \frac{4\pi}{Z} \int_0^\infty \rho(r) e^{-\alpha r} r^{k+2}\, dr,$$

and

$$\delta R_k = C_Z \delta E, \tag{2}$$

which follows from first-order perturbation theory.

Taking $\alpha = 0.15$ from muonic x rays,[8,10] the value of k associated with the electronic $1s_{1/2}$ shift using the electron potential of Seltzer[4] is $k = 2.917$. The corresponding conversion factor is found to be $C_Z = 78.1 \times 10^{-6}$ fm/meV. Since the optical IS is sensitive to the same R_k moment,[4] we convert the optical $\delta\langle r^2\rangle$ values[1] (which include a 10% uncertainty due to atomic Hartree-Fock calculations) using the conversion factor $\delta R_k = 0..1226\,\delta\langle r^2\rangle$ fm^{-1}. The parameters for the muonic case[8] are $k = 2.413$, and $C_Z = 1.328 \times 10^{-3}$ fm/keV. The results for δR_k for the three cases are shown in Table III. It is interesting to illustrate the comparison of the δR_k values associated with the IS for electronic 1s level, optical transitions, and muonic 1s levels with the help of Fig. 1. The drawn line is a calculated value[8] assuming a Fermi distribution. We note that the results from these different experiments are consistent within the quoted errors. A more detailed discussion of the muonic data will be given by the authors in Ref. 7.

The present electronic IS data may be used to normalize the optical IS directly (since the specific mass shift of these particular optical transitions is zero[1,2]). Using a least-squares fitting procedure, we calculate

$$\delta\nu = -55.1(3.1)\ \text{GHz/fm}^2\,\delta\langle r^2\rangle;$$

this differs from the factor $-44.9(5.0)$ GHz/fm^2 used previously[1,2] by 23% and amounts to multiplying the $\delta\langle r^2\rangle$ values quoted in Table III and taken from Ref. 1 by a factor 1.227(69).

A comparison of the experimental $\delta\langle r^2\rangle$ values with predictions from nuclear Hartree-Fock theory must await detailed calculations.

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