Energies of the muonic L and M transitions of the even-A Pb isotopes

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The energies of the muonic $4f \rightarrow 3d$ and $3d \rightarrow 2p$ transitions have been measured in the even-A lead isotopes, with several times the precision of any previous measurement. The isotope shifts and intradoublet energy differences are in excellent agreement with earlier work but the absolute energies generally differ by two or more standard deviations. The new results do not completely resolve the discrepancy between experimental and theoretical binding energies in muonic ^{208}Pb.

I. INTRODUCTION

The doubly magic nucleus ^{208}Pb is sufficiently well understood that one might expect the properties of the ^{208}Pb muonic atom to be adequately described by current quantum electrodynamic, nuclear polarization, and charge distribution models. However, Rinker and Speth[1] pointed out several years ago that there appeared to be a significant discrepancy between the calculated and experimental values of the energy splitting of the muonic $2p$ levels ($\Delta 2p$) of ^{208}Pb. Similar discrepancies seemed also to exist both in other muonic transitions (i.e., the $4f \rightarrow 3d$) and in other Pb isotopes (i.e., ^{204}Pb and ^{206}Pb). This early evidence was based on calculations that used a Fermi model for the nuclear charge distribution, and it was not clear to what extent the limitations of this model were responsible for the apparent discrepancies. The more recent study of Yamazaki *et al.*[2] used a model-independent combined analysis of muonic data[3] and electron scattering data;[4] it showed that the discrepancy was not an artifact created by the use of the Fermi model charge distribution. These authors took the experimental results at face value and interpreted the discrepancy in terms of a defect in the most uncertain muonic-atom correction, namely nuclear polarization. With this approach they were able to define an experimentally permissible range of values for the magnitude of the nuclear polarization effect in ^{208}Pb. The validity of their conclusions hinges, of course, upon the accuracy of the experiments.

In order to clarify the experimental situation regarding the muonic atom data and perhaps resolve the discrepancies, we have remeasured the energies of the muonic $4f \rightarrow 3d$ (M) and $3d \rightarrow 2p$ (L) lines of ^{204,206,208}Pb.

II. EXPERIMENT

The muonic x rays of ^{204,206,208}Pb were studied at the stopped muon channel of the Los Alamos Meson Physics Facility. The target masses and isotopic compositions are given in Table I. The data acquisition system consisted of a 60 cm^3 true-coaxial Ge(Li) detector and a highly stable (temperature regulated) and linear electronic system that has been described previously.[5,6] To minimize systematic errors in the isotope shifts, spectra from the three lead isotopes were collected simultaneously using a counter telescope that identified the particular target in which each muon stop occurred. The isotopically separated target samples were interchanged among the three possible target positions to reduce geometrical effects in the detected energies. Energy calibration was provided by spectra from γ-ray calibration sources that were stored simultaneously with the muonic x-ray data. Spectra obtained from ^{208}Pb in one of several runs are shown in Figs. 1 and 2.

The muonic spectra were fit with a function composed of a Gaussian convoluted with a Lorentzian with exponential tails. The tail parameters were determined from the γ-ray calibration lines and were held constant during fits of the muonic lines. The Lorentzian widths of the x-ray lines, which are quite evident in an element as heavy as Pb, were fixed at the computed natural linewidths. The energies were determined by linear interpolation from the calibration sources ^{24}Na, ^{88}Y, and ^{46}Sc and were corrected for detector system nonlinearity, which was determined in a separate measurement as discussed below.

In the case of ^{204}Pb and ^{208}Pb, the isotopic purity of our

TABLE I. Masses and isotopic compositions of the lead targets.

Target	% composition				Mass
	204	206	207	208	(g)
^{204}Pb	73.60	11.99	5.97	8.40	2.5
^{206}Pb	<0.001	99.984	0.003	0.012	9
^{208}Pb	<0.05	0.28	1.03	98.69	20

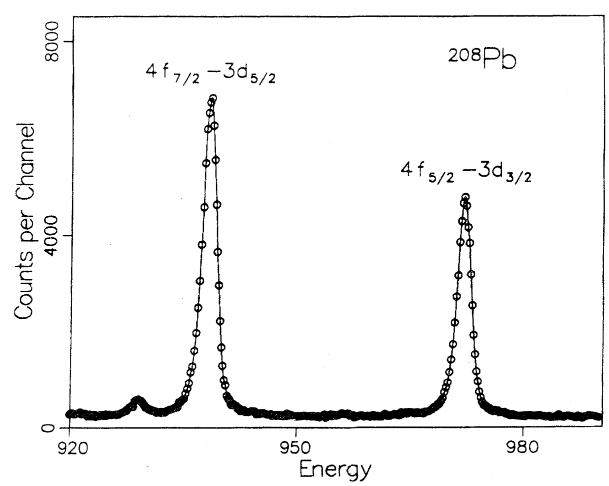

FIG. 1. Spectrum of the electromagnetic radiation of muonic ^{208}Pb in the 900-keV region.

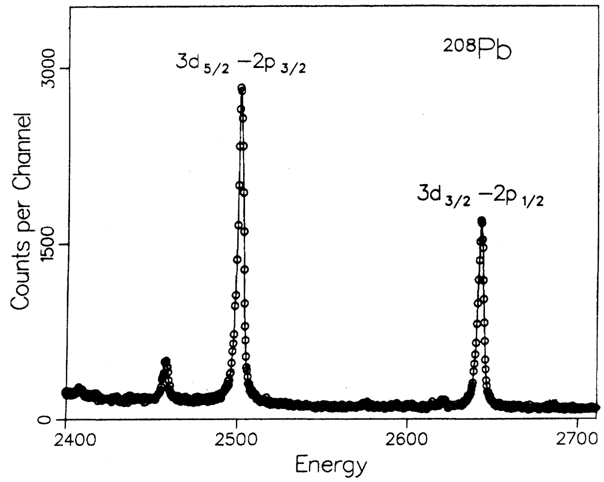

FIG. 2. Spectrum of the electromagnetic radiation of muonic ^{208}Pb in the 2500-keV region.

TABLE II. Energies of γ-ray sources used for calibration and evaluation of detector system nonlinearity.

Source	Energy (keV)	Reference
^{46}Sc	889.277 (3)	11
^{72}Ga	894.261 (5)	12
^{88}Y	898.042 (4)	11
^{110}Ag^{m}	937.493 (4)	10
^{72}Ga	970.702 (9)	12
^{72}Ga	999.918 (13)	12
^{72}Ga	1050.754 (6)	12
^{46}Sc	1120.545 (4)	11
^{24}Na	1368.633 (6)	10
^{88}Y	1836.063 (13)	10
^{72}Ga	1861.021 (13)	12
^{72}Ga	2490.986 (19)	12
^{72}Ga	2507.665 (20)	12
^{56}Mn	2522.943 (35)	12
^{56}Co	2598.460 (10)	10
^{24}Na	2754.030 (14)	10

targets (Table I) was not sufficient to permit the lines from other isotopes to be neglected. In these cases the fits included fixed impurity lines of appropriate relative intensities, with centroids determined iteratively from the present data (for the even-*A* isotopes) or computed from published isotope shifts[7] (for the odd-*A* isotope).

A. Background

The data were carefully studied for background lines that might perturb the apparent energies of the lead x-ray transitions. As expected, background near the *M* lines was observed due to muonic excitation of nuclear states in ^{204}Pb and in ^{207}Pb. In a run without lead targets and with no calibration sources, a weak background line at about 898 keV was observed. A background line near this energy is significant since it could alter the apparent energy of the 898-keV ^{88}Y calibration line. An examination of these data and other muonic data collected in previous experiments indicated that the most likely candidate for the source of the 898-keV line was ^{57}Fe. Other lines in the spectrum, e.g., 352, 1019, 1260, 1613, and 1724 keV, had intensities and energies[8] characteristic of thermal neutron capture on iron. The observed relative intensities of the 898-keV and these other transitions can readily be understood on the basis of an appropriate combination of thermal and resonant (1.167 keV) capture[9] of accelerator-produced neutrons in the iron of nearby magnets. The contributions of the contaminants (^{204}Pb, ^{207}Pb, and ^{57}Fe) to the apparent energy of the ^{88}Y line were estimated by making several fits to the spectrum in which the energies and intensities of the contaminating lines were varied within the limits imposed by our assumptions concerning their origins. A difference of 15 eV in the energy of the 898-keV calibration line could be so introduced.

In view of our uncertainty in the apparent energy of the ^{88}Y line, a separate measurement of ^{208}Pb used ^{46}Sc as a calibration source in place of ^{88}Y. Comparison of the energies of the $4f \rightarrow 3d$ transitions of the two separate data sets showed a maximum difference of 4 eV.

B. Nonlinearity

Detector system nonlinearity corrections were determined from ^{24}Na, ^{88}Y, ^{110}Ag^{m}, and ^{72}Ga for the *M* lines (energies of 937—972 keV) and ^{24}Na, ^{88}Y, ^{56}Mn, ^{56}Co, and ^{72}Ga for the *L* lines (energies of 2500—2645 keV). Energies for the calibration sources were taken from the recent evaluations of the Idaho[10,11] and Livermore[12] groups. A comparison of the energies of lines common to both evaluations indicated agreement at the level of a few eV, and we have therefore assumed that, for the present purpose, energies can be taken as necessary from either evaluation. The energies used in the present work are listed in Table II. The nonlinearity corrections at the energies of the lead lines were taken directly from the nearest γ-ray lines; for the *M* lines, the γ lines are within 2 keV of the muonic lines, and for the *L* lines they are within 8 and 45 keV. (A smooth curve fitted to all nonlinearity calibration data predicts a nonlinearity correction that is within 10 eV of the correction obtained by using only the nearest calibration line.) The nonlinearity of the electronic system was also checked using a precision (16-bit accuracy) computer-controlled pulser; the results were in good agreement with the γ-ray calibration source method.

C. Systematic errors

Several possible sources of systematic errors were considered. Data runs were made at three different amplifier gain settings to minimize differential nonlinearity effects. The results from all runs were mutually consistent and were combined to yield the final result. The γ-ray calibration data were stored simultaneously with the muonic x-ray data, using a technique which stores calibration events at a rate that is proportional to the instantaneous muon stopping rate. This technique reduces the possibility that slight electronic drifts could introduce a disparity between the muonic transitions and the calibration lines during runs that must take place over periods of several

TABLE III. Energies of muonic $4f\rightarrow3d$ and $3d\rightarrow2p$ lines in the even-A lead isotopes as measured in the present and previous experiments. The two errors listed for the present experiment are discussed in Sec. II D.

Isotope	Transition	Present experiment (keV)	Kessler *et al.* Ref. 3 (keV)	Jenkins *et al.* Ref. 14 (keV)	Martin Ref. 15 (keV)
^{208}Pb	$4f_{7/2}$-$3d_{5/2}$	938.111 (0.003)(0.013)	937.98 (0.06)	938.17 (0.15)	
	$4f_{5/2}$-$3d_{3/2}$	971.969 (0.005)(0.016)	971.85 (0.06)	971.99 (0.09)	
	$3d_{5/2}$-$2p_{3/2}$	2500.564 (0.010)(0.028)	2500.33 (0.06)	2500.34 (0.19)	
	$3d_{3/2}$-$2p_{1/2}$	2642.274 (0.012)(0.023)	2642.11 (0.06)	2641.94 (0.20)	
^{206}Pb	$4f_{7/2}$-$3d_{5/2}$	938.012 (0.009)(0.015)	937.87 (0.06)		937.9 (0.3)
	$4f_{5/2}$-$3d_{3/2}$	971.979 (0.015)(0.021)	971.86 (0.06)		971.9 (0.3)
	$3d_{5/2}$-$2p_{3/2}$	2501.812 (0.027)(0.038)	2501.60 (0.06)		2501.60 (0.20)
	$3d_{3/2}$-$2p_{1/2}$	2644.223 (0.045)(0.049)	2644.14 (0.06)		2644.50 (0.40)
^{204}Pb	$4f_{7/2}$-$3d_{5/2}$	938.120 (0.020)(0.023)	937.99 (0.06)		
	$4f_{5/2}$-$3d_{3/2}$	971.960 (0.027)(0.031)	971.86 (0.06)		
	$3d_{5/2}$-$2p_{3/2}$	2502.97 (0.12)(0.13)	2502.83 (0.06)		
	$3d_{3/2}$-$2p_{1/2}$	2645.66 (0.13)(0.14)	2645.52 (0.07)		

hours with varying accelerator beam intensities.

Geometry-related energy shifts in the Ge(Li) detector can result from a correlation between incoming photon direction and apparent energy.[13] To reduce these effects, the γ-ray calibration sources were placed behind, and approximately in line with, the lead targets as viewed by the Ge(Li) detector. This placement of the calibration sources, while maintaining the same incoming direction for both x rays and calibration-line photons, introduces the possibility of an apparent calibration energy shift due to shadowing of the detector face by the lead targets. To investigate the possibility of a shadowing effect, a run with the targets in place was compared with one without targets (a ^{56}Co γ-ray source placed off axis was used in both runs as a calibration reference). The data showed an upward shift of 6 ± 8 eV at 900 keV when the targets were removed. We have included a systematic error of 8 eV for possible geometric effects.

A shift in apparent energy of the muonic x-ray data with respect to the γ-ray calibration lines could also occur if there existed a small coupling of the muon telescope logic signals into the linear (energy measuring) electronics channel. Such a coupling could cause the γ-ray data, which are stored without scintillator telescope activity, to be shifted in energy with respect to the muonic x-ray data. In designing the experiment, we have attempted to reduce the electrical coupling between the logic and linear sections of the electronics as much as possible by physical isolation of the linear circuitry, by using multiply shielded coaxial cables where parallel cable runs were unavoidable, and by elimination of common ground return paths. A series of studies that used a precision pulser and simulated telescope logic signals indicated that any residual coupling was entirely negligible.

A further test of telescope-related energy shifts was made by simultaneously recording the muonic spectra from two ^{208}Pb targets, one of which was placed in the customary target position, the other placed immediately outside the counter telescope but within the region of the stopping muon flux. Since x rays from the second target were not coincident with counter telescope signals, they were stored as "calibration" events. The shift between the two spectra (for the 435-keV $5g\rightarrow4f$ x-ray lines) was found to be 1 ± 5 eV. In view of this investigation, we have included a systematic error of 5 eV due to a possible electrical coupling effect.

A systematic shift in energy between the x rays (which are coincidence gated) and the calibration lines (which are randomly gated) could also occur due to energy dependence of the Ge(Li) timing pick-off circuitry. Thus, a narrow coincidence timing window can slightly distort the observed line shape and produce a shift in apparent energy. In the present experiment a wide timing window was used to minimize the effect. No additional systematic error was included for time-window effects since the two-target ^{208}Pb test discussed above would reveal any residual effect of this type.

D. Results and error summary

The results of our measurements are listed in Table III. Two types of errors are given. The first is the error that

TABLE IV. Intradoublet energy differences and isotope shifts for the even-A lead isotopes. Because of the small energy separations involved, only "statistical" errors are listed for the isotope shifts; total errors are listed for the intradoublet transitions.

Isotope	Parameter or transition	Present experiment (keV)	Kessler *et al.* Ref. 3 (keV)	Jenkins *et al.* Ref. 14 (keV)	Martin Ref. 15 (keV)
^{208}Pb	$\Delta 3d$-$\Delta 4f$	33.858 (0.021)	33.87 (0.08)[a]	33.82 (0.17)[a]	
	$\Delta 2p$-$\Delta 3d$	141.710 (0.036)	141.77 (0.06)	141.60 (0.28)[a]	
^{206}Pb	$\Delta 3d$-$\Delta 4f$	33.967 (0.026)	33.99 (0.08)[a]		34.0 (0.4)[a]
	$\Delta 2p$-$\Delta 3d$	142.411 (0.062)	142.53 (0.05)		142.90 (0.45)[a]
^{204}Pb	$\Delta 3d$-$\Delta 4f$	33.840 (0.039)	33.87 (0.08)[a]		
	$\Delta 2p$-$\Delta 3d$	142.70 (0.19)	142.68 (0.08)		
$^{208-206}$Pb	$3d_{5/2}$-$2p_{3/2}$	1.25 (0.03)	1.27 (0.08)[a]		
	$3d_{3/2}$-$2p_{1/2}$	1.95 (0.05)	2.03 (0.08)[a]		
$^{206-204}$Pb	$3d_{5/2}$-$2p_{3/2}$	1.16 (0.12)	1.23 (0.08)[a]		
	$3d_{3/2}$-$2p_{1/2}$	1.44 (0.14)	1.38 (0.09)[a]		

[a]Computed from measured energies.

is relevant for computation of isotope shifts; it consists of a quadratic sum of the statistical measurement errors of each x-ray line and statistical uncertainties of the calibration and nonlinearity lines. A total error is also listed; it consists of a quadratic addition of the errors just mentioned, the absolute uncertainties of the calibration lines, the absolute uncertainty of our nonlinearity correction, and finally our estimate of the possible systematic errors discussed in Sec. II C. The results of several previous measurements[14,15] are also listed for comparison in Table III. Isotope shifts and intradoublet differences are given in Table IV.

III. DISCUSSION

The previous measurement of the lead L and M x-ray energies with the smallest quoted uncertainties is that of Kessler *et al.*[3] Compared to that measurement the present results are typically 2 to 5 times more precise. The present values are also systematically higher in energy than those of Ref. 3, by about 120 eV for the M x rays and by about 200 eV for the L x rays. In general, the two sets of values differ by significantly more than their quoted errors. Since γ-ray calibration energies used in the 1975 work of Kessler *et al.* differ somewhat from the more recent values used in the present work, we have, in the spirit of understanding the inconsistency, also analyzed our data using the older calibration energies used by Kessler *et al.* This procedure results, for the L x rays, in further increasing the difference by about 70 eV; for the M x rays, the difference is reduced, but only by about 10 eV. Whatever the origin of the inconsistency, it occurs only for the absolute energies, since the isotope shifts and intradoublet energy differences (Table IV) are in excellent agreement.

The absolute energies measured in the present experiment for the $4f \rightarrow 3d$ transitions, in contrast to the experimental results of Ref. 3, are in excellent agreement with the theoretical values (971.96 and 938.12 keV) of Rinker and Speth,[1] thus eliminating the discrepancy in the energies of the outer muonic transitions mentioned in the Introduction. However, since the discrepancy in the energies of the lower muonic levels in Pb rests principally on energy differences (especially $\Delta 2p$), these problems remain; they may be due to inadequate nuclear polarization corrections or to muonic-nuclear resonance excitation, as suggested in Refs. 1 and 2. In fact, there is growing evidence that discrepancies exist with $2p$-state nuclear polarization calculations in other regions of the periodic chart, for example,[16,17] ^{194}Pt, ^{150}Sm, ^{140}Ce, and ^{90}Zr. Clearly a fresh look at nuclear polarization (and perhaps quantum electrodynamic[18]) corrections in muonic atoms is now warranted.

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