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MUONIC X RAYS IN Pb^{206} AND POSSIBLE OBSERVATION OF NUCLEAR POLARIZATION*

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Muonic x rays in Pb^{206} were measured with high precision and in addition two of the weak transitions involving the $2s$ state were identified and measured for the first time. The new constraints make it difficult to fit the data with Dirac energies calculated for a finite nucleus and corrected for radiative effects. Consistency can, however, be achieved by depressing the calculated $1s$ energy by 6.8×2.3 keV, an effect tentatively interpreted as due to polarization of the nucleus by the muon.

In recent work at the Chicago cyclotron a special effort was made to measure the muonic x-ray spectrum of Pb^{206} with better statistics and higher precision than had been achieved previously.[1] Such measurements may be used to determine the size and shape of the nuclear charge distribution by comparison with the rather exact theory that is available for hydrogenlike atoms (Dirac equation), together with the applicable radiative corrections. An attempt is made to attribute remaining differences between theory and experiment to nuclear-polarization effects.

Pb^{206} was chosen because large targets of this element with high isotopic purity are readily available. Isotopically pure, but smaller, targets of Pb^{207} and Pb^{208} were also studied to determine the isotope shift. That work will be described in an extensive paper.[2]

Measurements were made with a 17-cm^3 pure coaxial-type Li-drifted Ge diode. The system had a resolution of 2.5 keV at 100 keV, 4.5 keV at 1.3 MeV, and 8.0 keV at 5.0 MeV. Pulse-height analysis was provided with a 4096-channel analog-to-digital converter connected to a PDP-8/ASI-6040 computer combination. Digital stabilization and a scheme for continuous monitoring of the system variations served to achieve an accuracy of about one part in 10^4 over a data collection period of two months. Further experimental details can be found in a conference report by Anderson.[3]

In that report a preliminary analysis of the data was presented. The measured energies of the following transitions in Pb^{206} were used to determine the nuclear-shape parameters: the two $2p$-$1s$ transitions, the three $3d$-$2p$ transitions, and

the two stronger of the three $4f$-$3d$ transitions, a total of seven pieces of information. The x-ray transition energies were calculated by solving the Dirac equation numerically for a nucleus with a two-parameter Fermi shape of the form

$$\rho(r)=\rho_0\{1+\exp[n(r/c-1)]\}^{-1}. \quad (1)$$

Here c is the half-density radius and the second shape parameter n is related to the usual skin-thickness parameter t by the relation

$$t=4.394c/n. \quad (2)$$

The calculated Dirac energies were corrected for vacuum polarization, Lamb shift, and the anomalous magnetic moment of the muon. The two parameters c and n were varied for the best fit. For this best fit, χ^2 divided by the number of degrees of freedom turned out to be slightly less than 1 and the problem of determining the nuclear-shape parameters of Pb^{206} from muonic atom data seemed to be essentially solved. Additional attention to this problem, however, showed that the analysis could be extended.

The procedure outlined, which is customarily used, was altered as follows:

(1) In our work on titanium[4,5] we found the weak transitions from higher p states to the $2s$ level and noted that, because the $2s$ state is strongly affected by the nuclear shape,[6] such transitions should be included in the analysis whenever possible. The intensities of transitions involving the $2s$ state are in the heavy elements of the order of 1% of the intensity of the principal lines, which explains why they have so far escaped detection. However, we had a very large amount of Pb^{206} data and good resolution so that we were able to identify the $3p_{3/2}$-$2s_{1/2}$ and $2s_{1/2}$-$2p_{1/2}$ transitions and measure their energies with sufficient precision to justify their inclusion in the data to be fitted. This was not the case for our Pb^{207} and Pb^{208} data and we are therefore limiting our present discussion to Pb^{206}.

(2) In order to constrain the fit somewhat more than in our preliminary work, we made use of the fact that the fine-structure splittings are in general measured to a higher precision than the transition energies. We have therefore replaced a certain number of transition energies by an equal number of splittings. We ended up with the nine energies and splittings listed in Table I. In this table, Δp is the fine-structure splitting of the $K\alpha$ line. The $L\alpha$ line has three components with splittings Δd and $\Delta p-\Delta d$. The $M\alpha$ line has also three components, but only the two strong ones are well separated and their splitting is labeled $\Delta d-\Delta f$.

Table I. Analysis of the principal muonic x-ray lines in Pb^{206}.

(1) No.	(2) Transition or fs splitting	(3) E_{exp}(keV)	(4) δE	(5) ΔE	(6) E_{calc}	(7) E'_{calc}
1	$3d_{3/2}$-$2p_{1/2}$	2643.75	0.36	0.47	2644.56	2644.07
2	Δp	185.65	0.12	0.15	185.70	185.46
3	$\Delta p-\Delta d$	142.30	0.24	0.25	142.83	142.60
4	$3p_{3/2}$-$2s_{1/2}$	1507.93	0.80	0.82	1508.90	1507.64
5	$2s_{1/2}$-$2p_{1/2}$	1217.81	0.80	0.82	1216.24	1217.11
6	$2p_{1/2}$-$1s_{1/2}$	5788.33	0.48	0.93	5787.12	5781.54
7	$4f_{5/2}$-$3d_{3/2}$	971.74	0.20		971.86	971.85
8	Δd	43.47	0.44		42.87	42.87
9	$\Delta d-\Delta f$	33.98	0.10		33.68	33.67
	Energies fitted				1-6	1-5
	Number of parameters				2	2
	N = number of degrees of freedom				4	3
	χ^2/N				3.59	1.42
	r_0				1.1979±0.0009	1.1987±0.0006
	n				13.99±0.56	14.37±0.41
	c				6.692±0.025	6.715±0.019
	t				2.102±0.079	2.053±0.055

In Table I, we list the measured energies in column 3 along with the experimental error δE. Three more points of difference between our preliminary treatment and the present analysis are of secondary importance, but should be mentioned for the sake of completeness.

(3) The amount of data included in the present analysis has been increased, resulting in small differences between the energies quoted in our preliminary report[3] and the present work. These differences, however, never exceed a fraction of the quoted experimental error.

(4) The vacuum polarization correction was calculated to first order in α according to the theory of Glauber, Rarita, and Schwed.[7] We estimated[8] that the uncertainty in the theoretical prediction is of the order of 1% of the calculated value of the vacuum polarization and this uncertainty was folded in with the experimental error δE before the fit was made. The compound errors ΔE which were used in the fit appear in column 5 of Table I.

(5) Only six energies were used in the fit because it turns out that the f-d transition energy and the d splittings are insensitive to a change of the shape parameters over a wide range. The values of the latter three energies have, however, been calculated from the fit and can be checked for consistency.

When the fitting procedure is applied to the first six energies of Table I, one obtains the parameters and calculated values of the energy, E_{calc}, as shown in column 6 of the table. It is

seen that the fit is not very good, the χ^2 per degree of freedom being equal to 3.6. In order to try to improve the fit, we have also used a nuclear charge distribution of the Fermi type with a parabolic depression,

$$\rho(r)=\rho_0(1+wr^2/c^2)\{1+\exp[n(r/c-1)]\}^{-1}. \quad (3)$$

It turns out, however, that the fit is not improved by taking w different from zero. Similarly, we have not been able to improve the fit significantly by assuming the existence of a nuclear halo as suggested by Barrett et al.[9] While we cannot exclude the possibility that a charge distribution of different shape might produce a better fit, the search for such a charge distribution could hardly be made meaningful until a proper account of possible nuclear effects, especially the nuclear polarization, can be given. Corrections for the latter have not been made so far for lack of solid theoretical calculation. We therefore restrict the present discussion to the two-parameter charge distribution (1).

Since the nuclear-polarization effect is greatest when the muon is in the $1s$ state, a more reliable measure of the nuclear-shape parameters should be obtained by excluding the $2p$-$1s$ transitions from the sample to be fitted. This was done by fitting the energies 1 to 5 of Table I, with the result given in column 7. It is seen that χ^2 is now only 1.4 per degree of freedom. On the other hand, the calculated $2p_{1/2}$-$1s_{1/2}$ energy difference turns out to be 6.8 ± 2.3 keV lower than the experimental value. We assume, tentatively, that this difference is significant and is due to nuclear polarization.

The shifts due to nuclear polarization were recently calculated by Cole[10,11] and Chen[12] using the experimental inelastic electron scattering cross sections which contain the same nuclear electromagnetic transition vertex as the dispersion corrections to the muonic energy levels. Cole's calculations cover the range of elements from $Z=8$ to $Z=70$, taking into account average nuclear properties, and his results must be extrapolated to $Z=82$ in order to be applicable to our experiment on lead. Chen's calculations, on the other hand, refer specifically to Pb^{208}. Both results are in good agreement. The estimated nuclear polarization contribution to the $1s$ level energy is 5.7 keV (±50%) according to Cole and 6.0 keV (±30%) according to Chen. These results are compatible with the discrepancy we have found between the calculated and experimental value of the $K\alpha$ energy. Although this agreement is suggestive it is still too early to draw definite conclusions for two reasons. First, the present analysis is based on the use of a Fermi charge distribution and is therefore not as general as one would like. Second, if nuclear polarization is present to the extent predicted by the theoretical calculations, it should affect also the higher energy levels. If such a correction were included in the calculation, the predicted $K\alpha$ energy could be expected to be different from the value we have obtained. Ideally, one would like to fit the whole set of experimental data with calculated energies which include the corrections for nuclear polarization. This, however, does not make much sense as long as the theoretical uncertainties of the polarization corrections are much larger than the experimental errors. Our attempt to fit the data with Cole's or Chen's predictions was inconclusive.

The nuclear-shape parameters obtained in this experiment are as follows:

$$r_0=1.199\pm0.001\ (\pm0.001)\ \text{F},$$
$$t=2.1\pm0.1\ (\pm0.1)\ \text{F}. \quad (4)$$

Here, the first error is the measurement error and the second error, in parentheses, is an estimate of a possible systematic error introduced by the uncertainty in the treatment of the polarization correction. This estimate was arrived at by considering extreme values of the nuclear-shape parameters obtained in various approaches such as those leading to the results in columns 6 and 7 of Table I.

In conclusion, while it is still too early to consider this analysis as an actual measurement of nuclear polarization by the muon, it tends to show that the possibility of studying this elusive effect is just around the corner. It is also evident that a more accurate calculation of the polarization correction is required before we can test the model to the accuracy inherent in the experiment.

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ULTRASONIC ELECTRON-NUCLEAR DOUBLE RESONANCE OF $CaF_2:U^{4+}$

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We report the first ultrasonic, electron-paramagnetic–nuclear double-resonance experiment. This experiment was performed on $CaF_2:U^{4+}$, and the results indicate that the ultrasonic-paramagnetic-resonance absorption line is inhomogeneously broadened due to superhyperfine interaction. These results are compared with a theoretical model for superhyperfine interaction in $CaF_2:U^{4+}$.

Strong ultrasonic-paramagnetic-resonance (UPR) absorption lines have been reported for the non-Kramers U^{4+} ion in trigonal sites in CaF_2.[1] The corresponding EPR lines are relatively weak. Satellite components have been observed in both the EPR and UPR spectra and interpreted[2] on the basis of a theoretical model as superhyperfine structure arising from the interaction of the substitutional U^{4+} ion with its eight nearest neighbor F^{19} nuclei. The observed satellite components correspond to the transitions $|\Delta M_S| = 2$, $|\Delta M_I| = 1$, whereas the components corresponding to the transitions $|\Delta M_S| = 2$, $\Delta M_I = 0$ are completely unresolved. The results of this analysis suggested a double-resonance experiment similar to an electron-nuclear double-resonance (ENDOR) experiment in EPR. Such an ultrasonic electron-nuclear double-resonance, or UNDOR, experiment would be quite significant in that it is a technique for performing ENDOR-type experiments on paramagnetic crystals which have relatively weak EPR but strong UPR absorption. In this experiment we demonstrate for the first time the feasiblity of this technique and use it to study the superhyperfine structure which is expected to contribute to inhomogeneous broadening of the observed main UPR component, namely the $|\Delta M_S| = 2$, $\Delta M_I = 0$ transitions. The usual problems concerning the coil-cavity combination in ENDOR are not present in UNDOR, since an rf coil may be wound directly upon the crystal which is external to the UPR cavity without affecting the cavity Q in any manner.

We have observed changes in the UPR absorption as a function of the frequency of an applied rf signal. With fixed applied resonance magnetic field, the change in the absorption of ultrasonic waves was measured, as the frequency of the applied rf signal was varied, by observing the change in the height of an echo that resulted from one or more passes of the ultrasonic pulse through the sample. A conventional x-band UPR spectrometer[3] with a two-turn rf coil wrapped around the sample, which is outside the UPR cavity, was used for these experiments. The coil was positioned such that the rf field $\vec{H}_{rf}$ was always perpendicular to both the applied field $\vec{H}_0$ and the symmetry axis of the impurity site.

In this experiment $\frac{1}{4}$-μsec ultrasonic x-band pulses were generated at a repetition rate of 500 pps. The first echo was observed. In order to avoid undue heating of the rf coil which was in liquid helium, the rf signal was pulsed such that it was on just prior to and during the time of transit of the ultrasonic wave. We have used cw radio frequency in these experiments with no