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Isotope shifts at 1.28 μm in Pb

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The isotopic shift of the $6p^2\,{}^3P_0 \rightarrow 6p^2\,{}^3P_1$ (1.28 μm) magnetic-dipole transition in Pb, which had been previously estimated incorrectly in the literature, has been measured, together with the magnetic hyperfine constant of the ${}^3P_1$ level in ^{207}Pb. The shifts of the ^{206}Pb and ^{207}Pb lines from the ^{208}Pb position are $\delta(206\text{–}208)=220.4(15)$ MHz and $\delta(207\text{–}208)=138.0(17)$ MHz, and the hyperfine constant $A(6p^2\,{}^3P_1)=-2388.2(45)$ MHz. Inclusion of the shifts in calculated optical rotation line shapes removes long-standing discrepancies between theory and experiment, which have limited the precision of parity-nonconservation measurements in Pb.

I. INTRODUCTION

In an earlier measurement of parity-nonconserving optical rotation in atomic Pb [1], discrepancies between theory and experiment in absorption and Faraday-rotation spectra contributed substantial uncertainty to the result. These differences, initially attributed to laser spectral impurities, persisted in subsequent measurements with a clean laser profile. We came to suspect the problem lay with hidden isotope shifts in the spectrum of natural Pb at 1.28 μm that were much larger than had been claimed in the literature. We report here the results of optical isotope shift measurements on ^{206}Pb, ^{207}Pb, and ^{208}Pb that confirm our suspicions and from a theoretical standpoint fit in well with the known isotope shifts of other transitions in Pb. We also sketch the consequences for optical rotation line shapes and parity nonconservation.

II. METHOD

The line shifts were observed directly by absorption spectroscopy of enriched Pb isotopes purchased from Oak Ridge National Laboratory. The 5-mW output of a cleaved-coupled-cavity semiconductor laser [2] is split threefold and passed through vacuum vapor cells, each containing 3–5 mg of metal. The 2.5-cm diam by 15-cm quartz cells are heated to 990 °C, for approximately 20% attenuation on absorption line center. About 5% of the unsplit beam is diverted through an optical isolator to a flat-plate Fabry-Pérot étalon with 1 GHz free spectral range. All signals are detected by germanium photodiode–amplifiers, digitized, and averaged by a minicomputer. The minicomputer also generates a 4-Hz triangle wave which is added to the laser injection current; this tunes the laser frequency across the absorption line [2]. Forward (to higher frequency) and reverse sweeps are recorded separately and analyzed independently. Simultaneous acquisition of the four signals ensures that their frequency axes are identical. Thirty-two data runs (sixteen forward-reverse pairs) were obtained, each an average of 1000 sweeps.

III. ANALYSIS

The $6p^2\,{}^3P_0 \rightarrow 6p^2\,{}^3P_1$ line has a single component in ^{206}Pb and ^{208}Pb, which have zero nuclear spin, and two components in ^{207}Pb, where magnetic hyperfine interaction with the spin-$\frac{1}{2}$ nucleus splits the ${}^3P_1$ level into $F=\frac{1}{2},\frac{3}{2}$ levels in the ratio 2:1. Line positions are found by a nonlinear least-squares fit to the three absorption spectra for each run, and associated Fabry-Pérot transmission spectra are similarly fit to calibrate the common frequency axis and correct for small sweep nonlinearities. The theoretical spectra include small impurity lines according to the Oak Ridge assays: 1% ^{207}Pb in the ^{208}Pb spectrum, and 6% ^{208}Pb, 2.4% ^{206}Pb in the ^{207}Pb spectrum. All other impurities were below 0.3%; tests showed their omission did not affect the fitted line centers. The laser Lorentzian width (120 MHz) and vapor Doppler broadening (510 MHz) are included by convolution integral, while the natural and collisional widths are neglected for this case of a metastable excited state, weak absorption, and the absence of buffer gas.

Figure 1 shows the data and best-fit curves for the for-

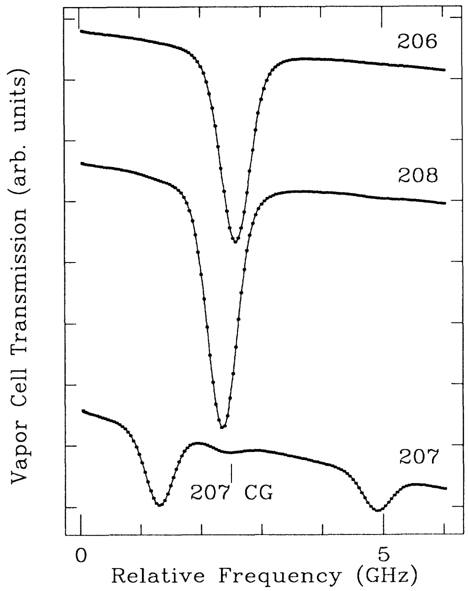

FIG. 1. Experimental isotope absorption spectra (solid circles) and computer fit (solid curves) at 1.28 μm in Pb. Spectra are identified by an atomic number at the right, and the ^{207}Pb center of gravity (CG) is indicated. Baselines are suppressed.

ward sweep of one run. The isotope spectra have been separately scaled and their baselines suppressed for ease of comparison. The background slope is due to a change in laser intensity proportional to the change in injection current, an unavoidable consequence of the frequency tuning. Component positions in ^{207}Pb are fitted independently, and the indicated center of gravity is then determined by the 2:1 splitting of the F states. Data were obtained with sweep widths ranging from 6 GHz, as shown in Fig. 1, to 8.5 GHz.

Results of the data analysis are summarized in Table I. The largest source of error, well resolved only in the $\delta(206-208)$ shift, is a systematic dependence of component separation on sweep width, with narrower sweeps producing larger values for the shifts and hyperfine constant. This is due to a thermal time constant of a few milliseconds in the laser crystal causing the laser frequency, which is determined by lasing stripe temperature, to lag the sweep current. This time lag distorts the frequency sweep near the edges (where the sweep reverses direction), and narrower sweeps are thus more susceptible. The fitting function for the Fabry-Pérot traces includes thermal effects, but the distortion is not completely removed. Although a wider frequency sweep reduces edge effects, sweeping beyond about 8.5 GHz decreased instrument resolution to the point where width effects at the same level could not be resolved. For the uncertainty due to this effect, we take as an upper limit the differences between narrow-sweep and wide-sweep average values; these are the systematic errors listed in Table I. The statistical errors reflect the scatter among runs, while the calibration errors account for uncertainties in both the Fabry-Pérot free spectral range and the frequency axis determination. Final values are the averages of all runs, with uncertainties obtained by combining the statistical and calibration errors in quadrature, then simply adding the systematic errors.

There are no direct determinations of the isotope shift in the 1.28-μm Pb line, but one indirect measurement has been made. Murakawa [3], using a hollow-cathode discharge and recording Fabry-Pérot fringes on photographic plates, measured the optical isotope shifts in 20 Pb lines. Among them were the transitions $6p7s\,^3P_1 \rightarrow 6p^2\,^3P_1$ (3640 Å) and $6p7s\,^3P_1 \rightarrow 6p^2\,^3P_0$ (2833 Å), which by their difference placed the shift in the 1.28-μm line at 30(40) MHz for both the $\delta(206-208)$ and $\delta(207-208)$ displacements. It is this erroneous result that initially led us to reject isotope shifts as the source of difficulty.

A more useful quantity for comparison with other work is the relative isotope shift (RIS), expressed in this paper as the $\delta(207-206)$ shift normalized to the $\delta(208-206)$ shift. In heavy atoms, isotope shifts are due almost entirely to nuclear charge distribution and volume effects [4], so the RIS is approximately independent of the particular transition in which the shifts are measured. That this is the case for Pb may be seen in Table II, which lists the RIS for a number of transitions in atomic and ionic Pb as compiled by Stroke [5]. It is interesting that the values demonstrate the odd-even staggering effect, where the odd-isotope center of gravity lies closest to the lighter even-isotope line.

TABLE I. Measured isotope shifts δ, hyperfine constant A, and associated errors for the 1.28-μm line in Pb. All values are in MHz.

	$\delta(206-208)$	$\delta(207-208)$	$A(6p^2\,^3P_1)$
Statistical error	0.2	0.6	0.7
Calibration error	0.2	0.1	1.9
Systematic error	1.2	1.1	2.5
Final value	220.4(15)	138.0(17)	−2388.2(45)

The hyperfine constant of the $6p^2\,^3P_1$ level in ^{207}Pb was measured by Mrozowski [6], who in 1940 reported $A(6p^2\,^3P_1) = -2380(20)$ MHz; the result in Table I is in good agreement.

IV. OPTICAL ROTATION LINE SHAPES

In our optical rotation measurements of parity nonconservation (PNC) [1], calibration of the apparatus rests on computer fits to two line shapes. The first is an absorption spectrum, which provides the optical depth of the vapor. The second is a Faraday rotation of linearly polarized light induced by applying a known magnetic field to the vapor along the light path, which together with the optical depth gives an absolute calibration of rotation signals. Thus the accuracy of a PNC measurement hinges on our understanding of these line shapes.

As displayed in Fig. 1, the isotope shifts are obvious. But in a natural Pb absorption spectrum, as in our PNC work, relative abundances and vapor widths mask the shifts. The only clues to their presence are a broadened even-isotope line and a shift ~60 MHz of the ^{207}Pb center of gravity relative to the combined even-isotope line center. Figures 2(a) and 2(b) illustrate this subtlety. An experimental spectrum with about 13 optical depths on line center has been fit (a) without isotope shifts and (b) including the measured shifts in Table I. The discrepancy in Fig. 2(a) is further obscured by the fitting process itself, which compensates by adjusting the composite line center, optical depth, and vapor widths away from their true values. The disagreement between theory and experiment is more apparent in the Faraday-rotation curves of

TABLE II. Measured relative isotope shifts (RIS) for transitions in atomic or ionic Pb, compiled by Stroke [5]. The RIS is the $\delta(207-206)$ shift normalized to the $\delta(208-206)$ shift.

Transition	RIS
$6p^2\,^3P_0 \rightarrow 6p^2\,^3P_1$	0.374(10) [a]
$6p^2\,^2P_0 \rightarrow 6p7s\,^3P_1$	0.374(11)
$6p^2\,^3P_0 \rightarrow 6p7p\,^3P_0$	0.375(10)
$6p^2\,^3P_2 \rightarrow 6p7s\,^3P_1$	0.376
$6p^2\,^1D_2 \rightarrow 6p6d\,^3D_2$	0.370
$6p^2\,^1S_0 \rightarrow 6p8s\,^3P_1$	0.372
$6s^2 6d\,^2D_{5/2} \rightarrow 6s^2 5f\,^2F_{7/2}$	0.377
$6s6p^2\,^2D_{5/2} \rightarrow 6s^2 7f\,^2F_{7/2}$	0.370
$6s6p^2\,^4P_{5/2} \rightarrow 6s^2 5f\,^2F_{7/2}$	0.382

[a] This work.

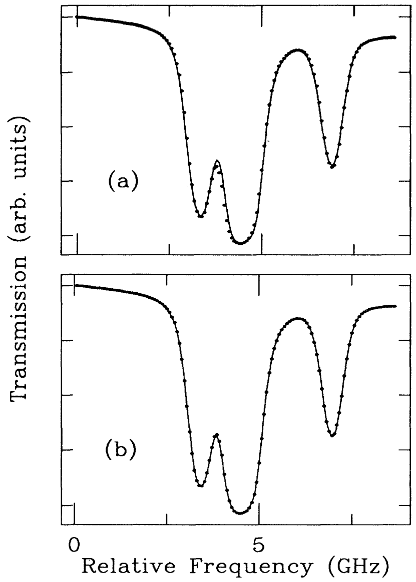

FIG. 2. Measured absorption spectrum (solid circles) of natural Pb at 1.28 μm. Solid curves are computer fits (a) without isotope shifts, and (b) with the isotope shifts from Table I.

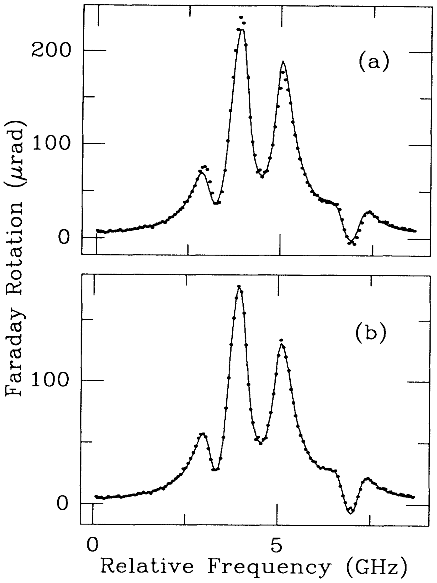

FIG. 3. Measured Faraday rotation (solid circles) corresponding to the absorption pattern of Fig. 2. Solid curves are computer fits (a) without isotope shifts, and (b) with the shifts from Table I.

Figs. 3(a) and 3(b), again fit (a) without shifts and (b) with the measured shifts. The experimental Faraday rotation was recorded along with the absorption of Fig. 2, and results from an axial field of 22 mG. Just as for the absorption, the fitting program has attempted to compensate for the shifts, yet the difference in height of the two rotation peaks about line center remains. It was the consistent failure of the theoretical curve to reproduce these peaks which led us to suspect a hidden isotope shift.

Figures 2(b) and 3(b) demonstrate that inclusion of the isotope shifts in theoretical absorption and Faraday spectra brings them into good agreement with measured line shapes. The question remains whether this improvement significantly alters our original PNC result for Pb. Because other systematic effects were present and the volume of data was large, a complete reanalysis is not practical. In addition, we expect a much more precise result from a modified apparatus. However, we are confident the change would be less than the quoted uncertainty of 25%, for the following reason: In fitting the absorption without knowledge of the isotope shifts, the most important error is an overestimation or underestimation of the optical depth, since rotations are proportional to this factor. The Faraday rotation is proportional to both the optical depth and the angle calibration factor, however, so the fit tends to compensate the optical depth error by underestimating or overestimating the angle calibration factor to get the proper overall scaling of the Faraday rotation. The parity rotation is proportional to the same product of factors, and thus the magnitude of measured PNC effects is relatively insensitive to the individual discrepancies. Fits to artificial parity data, using theoretical curves with and without isotope shifts, have produced differences of no more than 15% in the PNC results.

V. CONCLUSION

We have measured and corrected the isotope shift in Pb at 1.28 μm and have shown its importance in the calculation of absorption and Faraday-rotation line shapes for PNC measurements.

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