Hyperfine-structure measurements on trapped Pb II

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The $6P_{3/2}$-$6P_{1/2}$ magnetic dipole resonance transition in Pb^+ has been observed by cw laser excitation of an ion cloud stored in a Paul trap and subsequent detection of the fluorescence radiation. From the hyperfine-structure splitting of the spectrum we determine the A factor for the ground state, $A(P_{1/2})=12.967(13)$ GHz, and the excited state, $A(P_{3/2})=0.580(3)$ GHz. From a contamination of ^{208}Pb in our sample we derived the $^{207}Pb^+$-$^{208}Pb^+$ isotope shift [$\Delta\nu=311(14)$ MHz]. A small electric quadrupole admixture giving rise to a $\Delta F=2$ component in the spectrum has been determined to be 6.4(0.5)% of the total transition amplitude.

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INTRODUCTION

Optical measurements of the $6P_{1/2}$-$6P_{3/2}$ $M1$ fine-structure transition at 710 nm in lead ions by conventional spectroscopy [1] and by pulsed laser excitation in an ion trap [2] have results in values for the ground-state hyperfine-splitting (hfs) constant of the 207 mass isotope, which do not agree within the quoted limits of error. To clarify the situation we have performed narrow-band cw laser excitation on $^{207}Pb^+$ stored in a rf quadropole trap with proper calibration of the laser frequency. The experiment is also considered as a preparation for a laser-microwave double resonance experiment for a high precision determination of the ground-state hfs splitting in $^{207}Pb^+$. Such an experiment requires an uncertaincy of the optical hfs determination as low as possible, since the expected small signal rate in an $M1$-$M1$ double resonance experiment along with the narrow linewidth may make it difficult to find the microwave resonance. Preliminary results of this experiment have been published elsewhere [3].

EXPERIMENTAL SETUP

Isotopically enriched Pb^+ ions are created by surface ionization from a heated Pt filament placed near the inner surface of an ion-trap electrode. The Paul-type trap, made of stainless steel with hyperbolic shaped electrodes, has a ring diameter of 4 cm and was typically operated with 450-kHz driving frequeny of 600-V amplitude and a superimposed 10-V dc voltage. Holes of 6-mm diameter were drilled into the ring electrode to allow a laser beam to pass through the trap. One end cap consisted of a mesh of 60% transmission to observe fluorescence radiation from trapped ions after laser excitation. The trap was placed in an UHV vessel, whose base pressure was 1×10^{-9} mbar. We introduced He buffer gas into the vessel during operation to damp the ions' motion in the trap by collisions. Typical pressures range between 10^{-5} and 10^{-4} mbar. About 10^5 ions were stored simultaneously over periods as long as 14 days. The presence of ions in the trap could be monitored by their image currents induced in the trap's end-cap electrodes.

The laser system was a commercial linear cw dye laser, pumped by a 4-W Ar^+ laser. Single-mode operation with PP2 used as dye resulted in an output power of 70 mW. The laser frequency was stabilized by a temperature-controlled Fabry-Pérot étalon and the spectral bandwidth was 5 MHz. The laser wavelength was monitored by a commercial Michelson interferometer by comparison to a stabilized He-Ne laser. Laser scans were calibrated by the simultaneous observation of the transmission of a confocal étalon with 228.8-MHz free spectral range.

Laser excitation of the stored ion cloud was observed by counting fluorescence photons perpendicular to the laser beam. Since the lifetime of the excited $6P_{3/2}$ state was as long as 41.2 ms [4], the number of emitted photons per unit time was much smaller than the stray light from the laser, which mainly appeared at the entrance and exit windows of the apparatus. We therefore had to pulse the laser to separate the excitation and detection in time. Since the extinction of available electro-optical shutters was not large enough to avoid completely any stray light during the detection period, we chose mechanical shutters to open and block the laser beam. Usually the beam was on for 50 ms.

The optical detection system consisted of a cooled GaAs photomultiplier tube, a simple imaging system of two condenser lenses, and an interference filter. In order to avoid perturbation of the photomultiplier cathode by stray light during laser excitation we placed a mechanical shutter in front of the multiplier, which was opened for a time of 56 ms immediatelly following the laser excitation period. The total detection efficiency of the system was 10^{-4} including solid angle, transmission losses, and photomultiplier quantum efficiency. The rate of photomultiplier dark counts was 5 Hz, while the number of detected photons at resonant laser excitation was between 10 and 50 in one counting interval. Due to the time constants of the mechanical shutter the timing sequence could be repeated only twice per second. To illustrate the sensitivity of the system, we had not only to operate in a dark room, but also had to cover the red control lights of our electronic instruments to reduce the background counts to the required level.

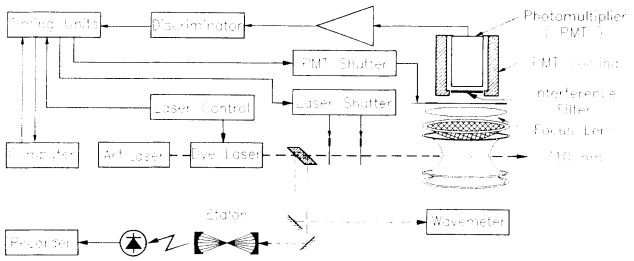

FIG. 1. Experimental setup.

The photomultiplier output was handled by a photon counting system and a subsequent personal computer, which served for data storage and timing control of the experiment. Figure 1 shows a block diagram of the experimental setup.

MEASUREMENTS

The laser was scanned over a 16-GHz range to cover the complete hyperfine spectrum of the 710-nm line. The total time for one scan was 2 min, which was repeated a few times for signal averaging. Figure 2 shows an example of such measurement, which exhibits the expected features from the level scheme: The lines labeled *a*, *b*, and *d* are the resonances from the $\Delta L=0$, $\Delta J=1$, and $\Delta F=0,\pm1$ magnetic dipole allowed transitions in $^{207}Pb^{+}$ and line *c* is an unsplit transition from a small contamination of $^{208}Pb^{+}$ in our sample. The linewidth of 1 GHz is determined by the Doppler width, which in our example corresponds to an ion temperature of 1600 K. The spectrum can satisfactorily be fitted by a sum of Gaussian functions.

The center wavelength of the spectrum was determined by a commercial wave meter to 710.172(2) nm which slightly improves tabulated values. To increase the resolution, in particular to resolve the excited-state fine-structure splitting between lines *a* and *b*, we reduced the trap's potential well depth, which results in a smaller average ion velocity at the expense of the stored ion number. Figure 3 shows a sum of ten scans over lines *a* and *b*. The linewidth is here reduced to 800 MHz corresponding to an ion temperature of 1027 K. Using the same trap potential for a scan of line *d*, we observe a shoulder at the high-energy side of the resonance (Fig. 4), which we attribute to a $\Delta F=2$ component in the spectrum, which is allowed by electric quadrupole radiation.

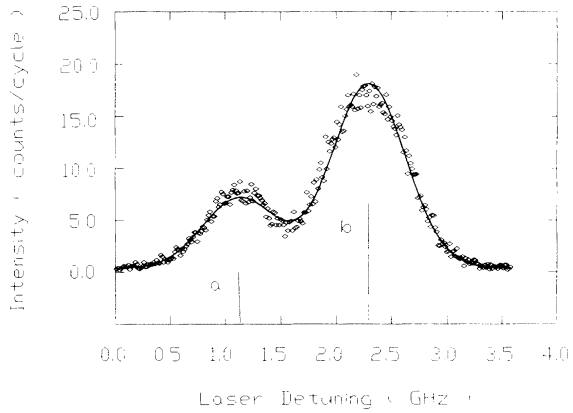

FIG. 3. Laser scan of the $F=1-F'=1$ (*a*) and $F=1-F'=2$ (*b*) hfs components in the $6P_{1/2}$-$6P_{3/2}$ fine-structure transition in $^{207}Pb^{+}$ at reduced potential depth. The linewidth corresponds to an ion temperature of 1027 K. Two Gaussian functions are fitted to the experimental points.

RESULTS AND DISCUSSION

From different spectra as shown in Figs. 2–4 we obtain information on the hyperfine spectrum of Pb^{+}. The fre-

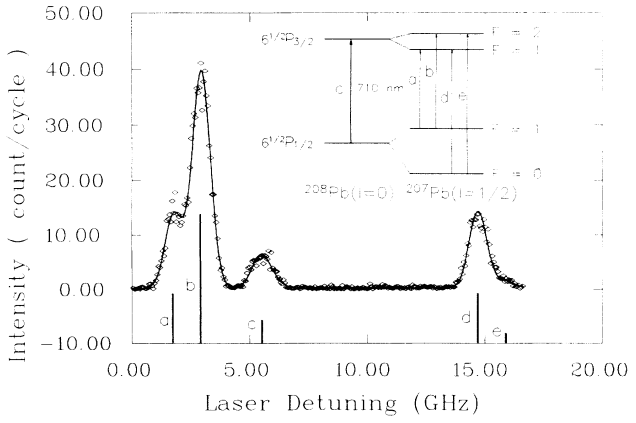

FIG. 2. Laser scan of the $6P_{3/2}$-$6P_{1/2}$ fine-structure transition in Pb^{+} at 710 nm. The resonances *a*, *b*, and *d* are hyperfine components from $^{207}Pb^{+}$ and *c* is from a contamination of $^{208}Pb^{+}$. The data are the average of ten consecutive scans. The Doppler limited linewidth corresponds to an ion temperature of 1600 K. The solid line consists of Gaussian functions fitted to the experimental points by a least-squares method.

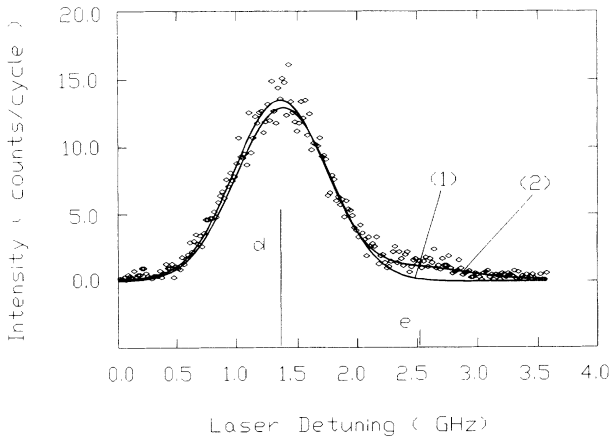

FIG. 4. Laser scan of the $F=0-F'=1$ hfs component in the $6P_{1/2}-6P_{3/2}$ fine-structure transition in $^{207}Pb^{+}$. From the comparison of one (1) and two (2) Gaussian functions fitted to the experimental points an additional component (*e*) is obvious, which we attribute to a weak admixture of an electric quadrupole allowed $F=0-F'=2$ radiation.

quency scale is given by the simultaneously recorded transmission maxima of a 228.8(1) MHz free-spectral-range Fabry-Pérot étalon, whose uncertainty does not contribute significantly to the errors quoted below in our results.

(a) $6P_{1/2}$ hyperfine splitting. The difference between lines *a* and *d* gives the ground-state hyperfine splitting. A fit by Gaussian functions gives a frequency difference of 12.967(13) GHz. The error is purely statistical. This result confirms the previous pulsed-dye-laser measurement [2] (12.85±0.10 GHz) but is in contradiction to the result of Ref. [1] (12.09±0.06 GHz). A comparison with theoretical expectations can be obtained by calculating the splitting from

$$A=\frac{\langle lj\|x^{(1)}\|lj\rangle}{[j(j+1)(2j+1)]^{1/2}}$$

which requires the knowledge of the reduced matrix element

$$\langle lj\|x^{(1)}\|lj\rangle=\frac{\mu_0 ce}{4\pi}\frac{\mu_I}{I}\frac{e\hbar}{2M_p}(-1)^l 2(2j+1)\sqrt{2}\times\begin{pmatrix} j & l & j \\ -\frac{1}{2} & l & -\frac{1}{2}\end{pmatrix}\int_0^\infty\frac{FG}{r^2}dr\ ,$$

where F and G are magnetic dipole radial hyperfine-splitting integrals.

Bouazza, Guern, and Bauche [1] have calculated $\int(FG/r^2)dr$ using radial Dirac-Fock functions and obtained 0.2933 for the $P_{1/2}$ stat. Taking $\mu_I=0.592\,583\mu_N$ [5] for the nuclear magnetic moment we obtain $A=12.22$ GHz, in fair agreement with the experiment.

(b) $6P_{3/2}$ hyperfine splitting. From the difference of lines *a* and *b* we obtain for the excited-state hfs splitting factor $A(6P_{3/2})=580(3)$ MHz compared to a previous optical determination of 500(30) MHz (Ref. [1]). The same theoretical consideration as above with $\int(FG/r^2)dr=0.0585$ from Ref. [1] gives $A=490$ MHz.

(c) Quadrupole transition. From a fit by two Gaussian functions to Fig. 4, taking the $P_{3/2}$ frequency splitting from Fig. 3, we obtain for the $\Delta F=2$ electric quadrupole transition an amplitude of (6.4±0.5)% of the total transition amplitude. Hults [6] reports a value of (3±1)% for the same amplitude.

(d) Isotope shift. The difference of the c.m. frequency of $^{207}Pb^+$ and line *c* from the $^{208}Pb^+$ contamination gives the isotope shift of the $6P_{1/2}$-$6P_{3/2}$ fine-structure transitions. We obtain a value of 311(14) MHz. A small part of it (5.3 MHz) is given by the normal mass shift, while the field shift

$$\delta\nu_{FS}=\frac{2\pi}{3}Z\alpha\hbar\Delta|\psi(0)|^2\delta\langle r^2\rangle$$

accounts for the rest. Fricke [7] has calculated the difference of the electron density $|\psi(0)|^2$ at the origin for the two states using relativistic multiconfiguration Dirac-Fock wave functions with extended nucleus and found 137.6 a.u. Using his result, we obtain a value of $\delta\langle r^2\rangle=-0.0693(32)$ fm^2 for the change in mean square radius, which is in good agreement with $\delta\langle r^2\rangle=-0.0723(24)$ fm^2 [8] obtained in neutral-atom spectroscopy for the same isotopes.

In conclusion, we have observed the magnetic dipole fine-structure transition on electromagnetically trapped Pb^+ isotopes and obtained a number of spectroscopic results. The experiment may be considered as a feasibility study of optical magnetic dipole hyperfine-structure transitions, which appear in highly charged single-electron systems around $Z=60$. Trapping of those ions at low energies and spectroscopic studies are presently under discussion [9].

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