High-precision hyperfine spectroscopy in $M1$-$M1$ double-resonance transitions on trapped $^{207}Pb^+$

X. Feng, G. Z. Li, and G. Werth
Institut für Physik, Universität Mainz, 6500 Mainz, Germany
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Optical pumping of the ground state of $^{207}Pb^+$ ions, confined in a Paul ion trap on the weak $6P_{1/2}$-$6P_{3/2}$ $M1$ transition at $\lambda=710$ nm, and consequent magnetic-dipole transitions between the ground-state hyperfine Zeeman levels have resulted in an A factor of 12 968 180 601.61(0.22) Hz and linewidth of 0.5 Hz. The value includes corrections of Zeeman shifts from residual magnetic fields and second-order Doppler shifts in the several-eV-deep trap potential, while the ions were cooled by He buffer gas. Two $M1$ quanta have been applied simultaneously in a double-resonance experiment.

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Precision hyperfine-splitting constants on trapped ions have been obtained in a number of cases [1–5]. Without exception the level structure of these ions is alkali-metal-like and strong $E1$ resonance transitions are available for effective optical pumping. The long observation times in ion traps along with the low relaxation rates between hyperfine levels under UHV conditions or at moderate buffer-gas pressures, however, allows optical pumping also under less favorable conditions.

We have investigated the spectrum of $^{207}Pb^+$. Laser excitation of a stored ion cloud on the $E1$–forbidden-$6P_{1/2}$-$6P_{3/2}$ resonance transition was reported previously and the lifetime of the excited state was determined to 41.2±0.7 ms [6]. Subsequently we have measured the hyperfine-splitting constants of ground and excited state by direct excitation of the $M1$ transition and a small $E2$ admixture with optical precision [7]. In this paper we present the results of an optical-microwave double-resonance experiment, where two $M1$ quanta have been used simultaneously. $6P_{1/2}$-$6P_{3/2}$-$M1$ excitation has been used for optical pumping and microwave-induced $M1$ hyperfine Zeeman transitions have been detected by a change in the intensity of the optical fluorescence.

A single-mode dye laser of 50-mW output power was tuned to one of the hyperfine components of the fine-structure line (see inset of Fig. 2) and depleted the corresponding ground level virtually completely with a time constant of 2 s. He buffer gas at 10^{-7} mbar led to relaxation from the other hyperfine level with a rate of 1.5 s. We used the buffer gas for moderate ion cooling by collisions. The temperature of the ion cloud of about 10^5 particles, determined from the Doppler width of the optical transition, was about 4000 K. We produced our ions by surface ionization from a Pt filament placed near one trap electrode and obtained storage times up to three weeks. Our trap of 2-cm radius was typically driven by a 450-kHz oscillator of 900-V amplitude and a superimposed 50 V dc voltage. The experimental setup is shown in Fig. 1.

We collected the fluorescence quanta from the excited ions through a mesh-trap electrode and condensor lens, which focused the light from the ion cloud to the photocathode of cooled GaAs photomultiplier tube. In order to avoid any stray quanta from the exciting light the laser was chopped mechanically. It was on for about 2000 ms for optical pumping and a shutter blocked the light path to the photomultiplier. Subsequently the ions interacted with a microwave field up to 1 s. We then probed the ground-state population in a second laser pulse of 100-ms duration. Afterwards we opened the photomultiplier shutter and counted the fluorescence quanta in a gate of 56 ms, corresponding roughly to the excited-state lifetime. This procedure was repeated every 4 s limited by the optical pumping time. The number of fluorescence counts during one counting interval was about 20 at resonance, while the dark counts were less than 1. The timing sequence is shown in Fig. 2.

The microwaves were produced in a klystron phase locked to the 20th harmonic of a quartz synthesizer,

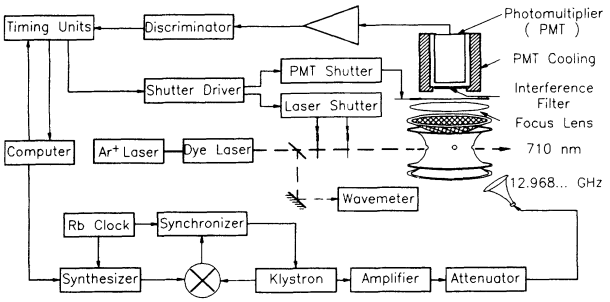

FIG. 1. Experimental setup.

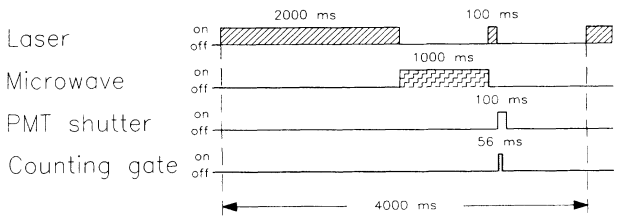

FIG. 2. Timing sequence.

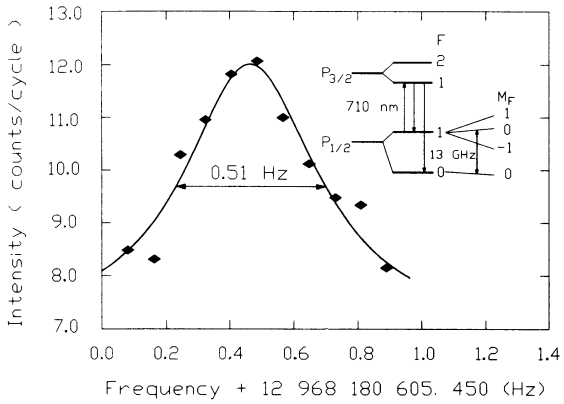

FIG. 3. $F=1, m=0$-$F=0, m=0$ ground-state hyperfine Zeeman transition in $^{207}Pb^{+}$. Each data point is the average of ten excitation-counting cycles, each lasting 4.0 s. The data are fitted by a Lorentzian line shape and a constant background (solid line).

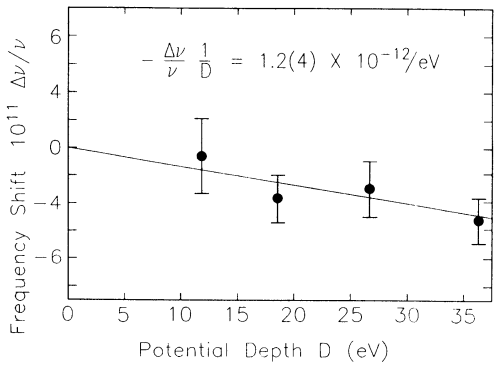

FIG. 4. Shift of the "magnetic-field-independent" hyperfine transition vs the trap potential depth. The extrapolation to zero depth corrects for the second-order Doppler shift.

which in turn was controlled by a Rb frequency standard and a broadcasted Cs-clock reference signal for short- and long-term stability, respectively. The absolute accuracy of the system is quoted to 1×10^{-12}. The microwaves were blown into the trap continuously by a horn antenna from outside the apparatus. When we blocked the microwaves while the laser was on to avoid light shifts, the transition frequency did not vary within the statistical error.

We observed microwave-resonance signals by an increase in fluorescence count rate. The observed lines could in all cases be well fitted by a Lorentzian line shape and the width was exclusively given by the microwave power. The minimum linewidth obtained in the "magnetic-field-independent" $F=0, m=0$-$F=1, m=0$ transition was 0.5 Hz (Fig. 3), limited by the frequency resolution of our present microwave system of 0.20 Hz. The data shown are the average of ten individual runs. The final results are taken from ten independent measurements as above.

We corrected the observed transition frequency for a small Zeeman shift from the residual magnetic field in the lab. The field was determined from the $\Delta F=1$ and $\Delta m=\pm1$ transitions, using the g_F factor from pure spin-orbit coupling. The corresponding shift amounts to 4.41 ± 0.03 Hz.

A second correction is due to a combination of second-order Doppler shift and Stark shift. We varied the trap potential depth by changing ac and dc amplitudes and extrapolated the observed frequencies to zero-field strength (Fig. 4). A linear extrapolation is appropriate, since the ion average kinetic energy is proportional to the trap potential depth [8,9]. For our typical operating point the shift is 0.57 ± 0.20 Hz. As a final result we have determined the ground-state hyperfine splitting of $^{207}Pb^{+}$ to 12 968 180 601.61 Hz.

The overall uncertainty is 0.22 Hz, which is the quadratic sum of uncertainties from statistics (0.088 Hz), Zeeman shift (0.03 Hz), and second-order Doppler shift (0.20 Hz). This improves previous optical determinations of the hyperfine splitting by about seven orders of magnitude [7,10]. We have performed a double-resonance experiment on trapped ions, where two $M1$ quanta have been used simultaneously. The experiment demonstrates the feasibility of precise hyperfine and fine-structure determinations even when there are no electric dipole transitions in the range of available lasers for optical pumping. Such cases can be found in doubly charged ions of Cs, Rb, and other ions of higher charge states.

The high precision obtained in this experiment and the prospects for further improvement may allow one to reconsider $^{207}Pb^{+}$ as a potential candidate for a frequency standard, as proposed earlier by Strumia [11]. The low rate of photon scattering on the $M1$ fine-structure transition certainly makes laser cooling nearly impossible, and other transitions are excluded because of their deep uv wavelength. If, however, another cooling mechanism like resistive cooling [12] is successfully employed—although the high mass of $^{207}Pb^{+}$ and the corresponding long-time constants for cooling are an obstacle, the ease of ion production and storage as well as the excitation wavelength, which may be produced by simple diode lasers, are of advantage—it could represent a compromise between physical properties and technical possibilities.

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