Ground State Hyperfine Splitting of Hydrogenlike $^{207}Pb^{81+}$ by Laser Excitation of a Bunched Ion Beam in the GSI Experimental Storage Ring

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Using a new bunched-beam technique in the GSI heavy-ion experimental storage ring (ESR), we performed precision laser spectroscopy on relativistic heavy ions in the hitherto inaccessible infrared optical region. We determined the wavelength of the $M1$ transition between the $F = 1$ ($\tau \approx 50$ ms) and $F = 0$ hyperfine states of the $1s$ ground state of hydrogenlike $^{207}Pb^{81+}$. Comparing the result of 1019.7(2) nm with very recent theoretical predictions concerning QED and nuclear size contributions, a disagreement of 4.5 nm is found. Since the nucleus of $^{207}Pb^{81+}$ is well described by the single-particle shell model, uncertainties in nuclear corrections are expected to be small. [S0031-9007(98)07624-8]

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The hyperfine splitting (HFS) of the $1s$ ground state of one-electron, two-body (hydrogenlike) system is the simplest and most basic magnetic interaction in atomic physics. In hydrogen the splitting is measured to thirteen significant figures, considerably more precise than the six-digit precision of the theoretical calculations of this quantity [1]. These calculations solve the Dirac equation and then add corrections for the effects of the finite size of the nuclear charge and magnetization as well as for the QED effects of self-energy and vacuum polarization. While the QED contributions are of the order of 10^{-6} to 10^{-5} for a single proton, these corrections are several percent in hydrogenlike ions of large Z in which the electron experiences exceptionally intense electric and magnetic fields. Thus measurements of the spectra of these systems can stringently test theoretical calculations of QED and nuclear effects.

Recently the $1s$ ground state transitions in high-Z, hydrogenlike ions have become accessible to optical spectroscopy at the experimental storage ring (ESR) at GSI-Darmstadt and at the electron beam ion trap Super-EBIT at Lawrence Livermore National Laboratory. Measurements of the ground state hyperfine splittings of $^{209}Bi^{82+}$ at GSI [2] and $^{165}Ho^{66+}$ at LLNL [3] have stimulated a large number of theoretical calculations of the wavelengths of these transitions [4–15]. Discrepancies are fond between theory and experiment for both $^{209}Bi^{82+}$ and $^{165}Ho^{66+}$.

The calculations for bismuth yield a value 1 nm (5×10^{-3}) larger than the measured value. On the basis of the precisions assigned to the corrections this discrepancy is significant, but corrections for the nuclear effects vary considerably depending upon how much the nuclear core is assumed to be polarized. For holmium, a smaller discrepancy between the calculated and measured values is reported [3], but the theoretical analysis did not take into account nuclear polarization [15] which is expected to contribute significantly.

In view of this unsatisfactory situation we measured the $1s$ ground state hyperfine transition of $^{207}Pb^{81+}$. We chose this nucleus because it is well described by the single-particle model. The magnetic moment has been measured with high precision in the atomic vapor phase using optical double resonance [16], which, although it remains necessary to correct for diamagnetic effects, avoids the ambiguity associated with the chemical environment in typical NMR measurements. Its nuclear magnetic moment is given very exactly by that of the $p_{1/2}$ neutron hole in the doubly magic nucleus ^{208}Pb (82 protons and 126 neutrons), and the effects of core polarization are expected to be less than 0.3%. Furthermore, precise measurements of the nuclear radius [17] of ^{207}Pb by means of electron scattering and studies of muonic x rays permit accurate calculation of effects due to the finite size of the nucleus.

Experiment.—Our experiment was performed using the ESR at GSI [18]. Natural lead contains 22.1% of ^{207}Pb, and this was selected by magnetic deflection from the ion source of the accelerator. Hydrogenlike lead ions were prepared from lower charge states by accelerating them in the heavy-ion synchrotron SIS and passing them through a stripping foil. At 200 MeV/nucleon, more than 20% of the ions end in the desired charge state. Up to 10^8 ions were accumulated in the ESR and cooled by Coulomb interaction with co-moving electrons in the ESR's electron cooler.

The $M1$ transition between the ground state hyperfine levels in hydrogenlike lead is in the infrared optical region at a wavelength of about 1 μm. As a consequence, the calculated lifetime of the upper hyperfine level is

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$\tau = 52$ ms. In order to study the spectroscopy of such an extremely long-lived infrared transition, it was important to develop a new technique of bunching the circulating ions.

This new technique has several advantages: Without bunching only a small fraction of the stored ions would be illuminated by the pulsed laser. With two bunches in the ring it is possible to illuminate 50% of the ions stored in the ring. It also becomes possible to compare fluorescence from a bunch containing excited ions with the light detected from a bunch containing no excited ions. Thus bunching both improves the efficiency with which ions are excited by the laser pulses and greatly facilitates background subtraction. The new technique improves the signal-to-background ratio by nearly a factor of 10. The experimental arrangement is shown schematically in Fig. 1. The circulating ions were compressed into two bunches by applying a radio-frequency acceleration voltage with an amplitude of 20 V. From the fact that it took each bunch about 60 ns to pass an observation point, the bunch size was determined to be about 11 m in length, i.e., about 10% of the ring circumference.

Although the wavelength in the rest frame of the ions is outside the range of efficient photocathode materials, standard near-IR photomultipliers sensitive up to about 800 nm can be used because the large velocity of ions circulating in the ESR Doppler shifts the wavelength of the fluorescence light by nearly a factor of 2. The storage ring is also essential because the long lifetime τ of the infrared transition of about 50 ms makes the fluorescence intensity rather low. In the storage ring, the beam has a storage time limited mostly by electron capture in the electron cooler, of about 20 min. The long lifetime of the beam also means that deexcitation by collisions, which could reduce the detectable fluorescence, must be very low. Consequently, a rather accurate measurement of the transition lifetime is possible.

To improve the collection of fluorescence photons, an array of cylindrical and elliptical mirrors was centered around the ion beam and arranged to produce a line focus on the photo cathode of three 2-in. photomultipliers (Hamamatsu R1017) selected for low background and high sensitivity to near-IR light. The mirrors were slightly tilted with respect to the beam direction to enhance the detection of light emitted into forward angles.

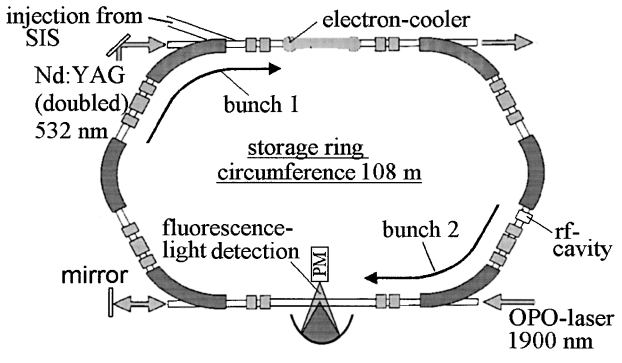

FIG. 1. The laser spectroscopy setup at the ESR. The circulating ions are bunched into two packets. Laser excitation is possible either by a parallel superimposed laser beam at 532 nm, or a counterpropagating beam at about 1900 nm. Excitation and detection are synchronized with the revolution of the ion packets.

The ESR also allowed us to use a fixed frequency Nd:YAG laser, doubled to 532.222(5) nm (pulse energy 250 mJ), to search for the resonance. Tuning was done by changing the beam velocity to vary the Doppler shift. The velocity of the ions was varied by changing both the acceleration voltage of the electron cooler and the buncher frequency. Although the velocity is affected mainly by the buncher frequency, a mismatch with the electron velocity would lead to a wider spread of the ion velocities. To minimize such broadening, we used electrostatic pickup signals to precisely measure the circulation frequency of the coasting beam at a given electron-cooler voltage, and then we adjusted the buncher frequency exactly to this value. As a result the precision of the determination of the ion velocity became the precision of the determination of the electron cooler's acceleration potential, which is known to be better than 10^{-4}.

The laser pulses were synchronized with the buncher frequency, so that only atoms in one of the two bunches were excited. Atoms were excited by the laser in one straight section of the ESR, and their fluorescence was detected in the other. The detection electronics was also synchronized with the bunches, so that photons detected during the passage of the different bunches were counted into different registers. Thus light from one bunch was beam related background only, and light from the other bunch was background plus signal. After the signal was found, an intracavity etalon was installed, narrowing the bandwidth to 0.005 nm. The wavelength was then measured to this precision relative to a calibrated single-mode He-Ne laser. The raw data was shown in Fig. 2, where the photon counts are plotted against the acceleration voltage applied at the electron cooler.

Typically, the data were recorded for about 30 min after each filling of the ESR, and during this time the ion-beam intensity decreased by a factor of 3. Therefore the counts of beam-dependent background (dotted line),

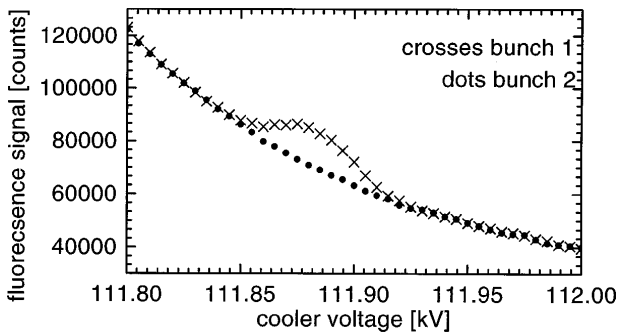

FIG. 2. Raw data from laser induced fluorescence: Dots represent the signal detected from the bunch without laser excitation, and crosses represent the signal including fluorescence.

mostly from ionization and excitation of residual gas, appear to decrease as the acceleration voltage is changed. The count rate in the bunch excited by the laser (crossed line) shows exactly the same behavior, but with the fluorescence signal added at resonance.

The background-corrected and normalized signal is shown in Fig. 3 as a function of the wavelength in the rest frame of the ions. This line position is in accordance with the value of the magnetic moment obtained by optical pumping double resonance [16], and rules out an older value determined by NMR and cited in Ref. [19]. The measured signal width of 0.1 nm is composed of several parts. The large contribution is due to the Doppler width $\Delta\lambda_0 = \lambda_0\beta\gamma^2\frac{\Delta\beta}{\beta}$ where $\Delta\beta/\beta = 2 \times 10^{-4}$. Mismatches between the buncher frequency and the velocity of cooler electrons increase linewidth as do Zeeman and Stark splittings, and the effect of the possible small angle between the laser beam and the ion beam. All of these, however, contribute much less than the Doppler width. The accuracy of the measurement is therefore, in principle, smaller than the width of the signal, but we have included the full width as an upper estimate of all possible systematic effects. The uncertainty of the ion-velocity determination of $\delta\beta/\beta = 1 \times 10^{-4}$ enters into the final result upon transformation into the rest frame of the ions. The centroid of the resonance peak corresponds to 1019.7(2) nm.

It is possible to determine the ion velocity more accurately by also measuring the hyperfine transition with a laser beam directed opposite to the ion beam. We used 1900 nm light from a Nd:YAG-pumped optical-parametric-oscillator (OPO) system to take a rough spectrum which verified the value of 1019.7 nm.

A fit of the exponential decay of the fluorescence after excitation by a pulse shows that the mean decay time is 49.5(6.5) ms in the ions' rest frame. This is about 5% less than predicted, but agrees with theory within the relatively large statistical uncertainty of 6.5 ms.

Table I shows theoretically calculated and experimentally obtained wavelengths for the ground state hyperfine transitions of hydrogenlike lead and hydrogenlike bismuth. The experimental values are both 0.5% smaller than the corresponding calculated values.

To discuss possible sources of these discrepancies it is convenient to view the ground state hyperfine splitting of hydrogenlike ions as a product of the nonrelativistic solution multiplied by correction factors

$$\Delta E(\mu) = \frac{4}{3}\,\alpha(\alpha Z)^3\,\frac{\mu}{\mu_N}\,\frac{m}{m_p}\,\frac{2I+1}{2I}\,mc^2 \times \{A(\alpha Z)(1-\delta)(1-\varepsilon) + x_{\mathrm{rad}}\}. \quad (1)$$

Here α is the fine structure constant, Z is the nuclear charge, m is the electron mass and m_p the proton mass, μ is the nuclear magnetic moment, μ_N is the nuclear magneton, I is the nuclear spin, and $A(\alpha Z)$ is the relativistic correction. The factor $(1-\delta)$ corrects for the finite spatial distribution of the nuclear charge

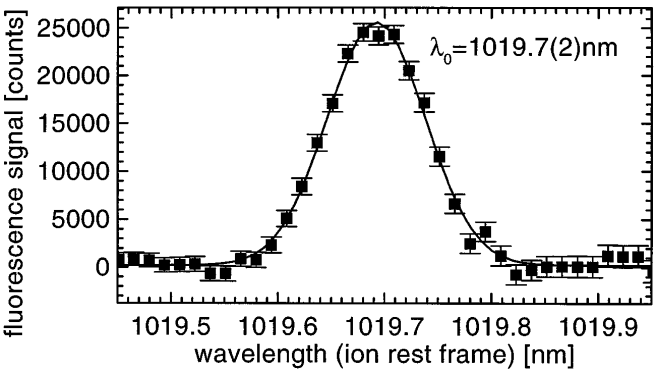

FIG. 3. Results obtained for hydrogenlike lead: Spectrum of laser induced fluorescence vs excitation wavelength. The signal width represents the Doppler width of the ion beam, whereas the given accuracy is due to the knowledge of the ion velocity in the storage ring.

(Breit-Schawlow correction), $(1-\varepsilon)$ corrects for the finite spatial distribution of the nuclear magnetization (Bohr-Weisskopf correction) [20], and x_{rad} is the QED correction. The sizes of the corrections given in Table I for $^{209}Bi^{82+}$ and $^{207}Pb^{81+}$ were obtained from Refs. [3], [9], and [10].

The fully relativistic calculation of the transition wavelength usually includes the Breit-Schawlow correction in a Dirac-Fock calculation. Even though there is slight model dependence of the nuclear radius, the various calculations are highly accurate and produce similar values. The uncertainties assigned to these nuclear corrections are ± 0.2 nm [10].

Calculations of corrections for the effect of the spatial extent of the nuclear magnetic moment, the Bohr-Weisskopf effect [20], are complicated by nuclear core polarization, and different approaches give different results. The smallest correction results from using the single-particle model [7], where the distribution of the magnetic moment is obtained from the wave function of

TABLE I. The different contributions to the hyperfine splitting.

	$^{209}Bi^{82+}$	$^{207}Pb^{81+}$
rms radius	5.519 fm	5.497 fm
Magnetic moment (corrected) [4,5,13]	4.1106(2)μ_N [19]	0.58219(2)μ_N [16]
Point nucleus (Dirac) [4,5,8]	212.320(1) nm	885.76(3) nm
+Breit-Schawlow	238.791(50) nm	989.66(10) nm
+Bohr-Weisskopf [10]	243.91(38) nm	1019.1(1.9)[0.4] nm
Vacuum Polarization [7]	−1.64 nm	−6.83 nm
Self-energy [11]	+2.86 nm	+11.9 nm
Total QED [11]	+1.22(10) nm	+5.08(50) nm
Theory incl. QED	245.13(58) nm	1024.2(2.4) nm
Experiment	243.87(1) nm	1019.7(2) nm

the $h_{9/2}$ proton for bismuth, or the $p_{1/2}$ neutron for lead. This seems inappropriate, especially in the case of bismuth for which the magnetic moment deviates by a factor of 2 from the single-particle estimate. The magnetic moment can be well reproduced for both cases by a dynamical mixing model [10]. For bismuth this approach yields a value for the Bohr-Weisskopf effect about 20% larger than the value from the single-particle model. In this case the calculation of the Bohr-Weisskopf effect can be checked independently by comparison with the measured ground state hyperfine splitting in the muonic atom [21], and agreement is excellent. In the case of lead, the contribution related to the dynamical mixing is much smaller, and the corrections calculated by the extreme single-particle approach [11] and the dynamical mixing procedure differ by only 0.25 nm. This is the direct consequence of the much simpler nuclear structure of lead which is well described by the single-particle shell model. We use the value from Ref. [9] and the new result computed for lead in Table I.

For $^{209}Bi^{82+}$ the uncertainty in the Bohr-Weisskopf contribution (see Table I) covers the confidence level manifested experimentally by the result in muonic bismuth [21]. For $^{207}Pb^{81+}$, on the other hand, the two extreme approaches agree within a fraction of this uncertainty. This is indicated by the uncertainty level of 0.4 nm in the square bracket. In view of the strongly different nuclear structure in the two cases, we believe that our new result considerably weakens the arguments that attribute the difference between experimental and theoretical values of the $1s$ hyperfine ground state transition wavelength to model-dependent uncertainties in the calculation of the Bohr-Weisskopf effect.

There remains then only two likely sources for the observed discrepancy, the value of the magnetic moment and the QED corrections. Different authors have evaluated QED corrections due to vacuum polarization and self-energy and obtained identical results [11,14,15] with estimated uncertainties of ± 0.05 nm. The percentage QED corrections are very similar for both elements.

Although the value of the magnetic moment of lead is measured to high precision, it must be corrected for diamagnetic shielding by atomic electrons. The commonly used value [19] of μ has been corrected using a shielding constant of 1.686%. This is the shielding constant calculated for doubly charged lead ions [22], despite the fact that μ was measured in neutral lead vapor.

From plausibility, the additional effect in the shielding upon adding just two more electrons should be small, and widely accepted calculations [23] assume the difference to be only 3×10^{-5} in literature. However, a conflicting shielding value of 2.055% for the neutral case exists [24]. If such an unexpected large change of nearly 0.4% in shielding would be effective, this could explain most of the discrepancy.

The new bunched-beam technique presented in this paper will allow in bismuth a direct comparison of the lithiumlike ($2s$) and the hydrogenlike ($1s$) HFS providing a QED determination mostly independent of the nuclear parameters.

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Note added in proof.—In a new article [M. G. H. Gustavsson and A.-M. Mårtensson-Pendrill, Phys. Rev. A **58**, 3611 (1998)] it is strongly suggested not to use the magnetic moment from Ref. [16]. Using the older value given in [19] a much better agreement between experiment and theory is reached.

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[1] S. G. Karshenboim, Phys. Lett. A **225**, 97 (1997).
[2] I. Klaft *et al.,* Phys. Rev. Lett. **73**, 2425 (1994).
[3] J. R. Crespo Lopez-Urrutia *et al.,* Phys. Rev. Lett. **77**, 826 (1996).
[4] M. Finkbeiner *et al.,* Phys. Lett. A **176**, 113 (1993).
[5] S. M. Schneider *et al.,* J. Phys. B **26**, L529 (1993).
[6] S. M. Schneider *et al.,* J. Phys. B **26**, L581 (1993).
[7] S. M. Schneider *et al.,* Phys. Rev. A **50**, 118 (1994).
[8] V. M. Shabaev, J. Phys. B **27**, 5825 (1994).
[9] L. N. Labzowsky *et al.,* Phys. Rev. A **51**, 4597 (1995).
[10] M. Tomaselli *et al.,* Phys. Rev. C **51**, 2989 (1995); M. Tomaselli *et al.,* Phys. Rev. C **58**, 1524 (1998).
[11] H. Persson *et al.,* Phys. Rev. Lett. **76**, 1433 (1996).
[12] V. M. Shabaev and V. A. Yerokhin, Pis'ma Zh. Eksp. Teor. Fiz. **63**, 309 (1996) [JETP Lett. **63**, 376 (1996)].
[13] T. Bastug *et al.,* Z. Phys. D **37**, 281 (1996).
[14] S. A. Blundell *et al.,* Phys. Rev. A **55**, 1857 (1997).
[15] V. M. Shabaev *et al.,* Phys. Rev. A **56**, 252 (1997).
[16] H. M. Gibbs and C. M. White, Phys. Rev. **188**, 180 (1969).
[17] H. De Vries *et al.,* At. Data Nucl. Data Tables **36**, 495 (1987).
[18] B. Franzke, Nucl. Instrum. Methods Phys. Res., Sect. B **24**, 18 (1987).
[19] P. Raghavan, At. Data Nucl. Data Tables **42**, 189 (1989).
[20] A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950).
[21] A. Rüetschi *et al.,* Nucl. Phys. **A422**, 461 (1984).
[22] F. D. Feiock and W. R. Johnson, Phys. Rev. **187**, 39 (1969).
[23] O. Lutz and G. Stricker, Phys. Lett. **35A**, 397 (1971).
[24] K. M. S. Saxena and P. T. Narasimhan, Int. J. Quantum Chem. **1**, 731 (1967).