# Hyperfine Splitting of the $2s_{1/2}$ and $2p_{1/2}$ Levels in Li- and Be-like Ions of $^{141}_{59}$Pr

P. Beiersdorfer,* E. Träbert,† G. V. Brown, J. Clementson, D. B. Thorn, M. H. Chen, and K. T. Cheng
*Lawrence Livermore National Laboratory, Livermore, California 94550-9234, USA*

J. Sapirstein
*University of Notre Dame, Notre Dame, Indiana 46556, USA*

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High-resolution spectroscopy of the $2s_{1/2} - 2p_{1/2}$ transition in the extreme ultraviolet region is shown to resolve the level splitting induced by the nuclear magnetic field of both the $2s_{1/2}$ and the $2p_{1/2}$ levels in lithiumlike $^{141}Pr^{56+}$ and of the $2s_{1/2}2p_{1/2}$ $^3P_1$ level in berylliumlike $^{141}Pr^{55+}$. The $^{141}$Pr ions are an ideal test of this measurement approach because their energy levels are known well from first principles and are unaffected by small energy contributions from QED and nuclear magnetization effects. The accuracy attained in the measured $196.5 \pm 1.2$ meV $2s_{1/2}$ splitting is more than an order of magnitude better than that achieved before using crystal spectroscopy of the $2s_{1/2} - 2p_{3/2}$ x-ray transition and at the level needed to implement a proposed scheme for disentangling the contributions from QED and nuclear magnetization effects in higher-$Z$ ions, such as $^{209}$Bi.

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The hyperfine splitting of the $1s$ ground state level leads to the famous 21 cm line of hydrogen. While this line is well understood for hydrogen, in part because of the development of the maser [1,2], it remains enigmatic for hydrogenlike ions of elements with a high atomic number $Z$. The first successful measurement of the $1s$ hyperfine splitting of a high-$Z$ ion focused on $^{209}Bi^{82+}$ [3]. It produced an energy value that could not be readily reproduced by theory [4–6], leading to speculation about the predictive capabilities of QED in strong magnetic fields. Moreover, the measurement of the radiative decay rate of the hyperfine transition could not be reproduced by theory. Subsequent measurements of the $1s$ hyperfine splitting of $^{165}Ho^{66+}$, $^{207}Pb^{81+}$, $^{185}Re^{74+}$, $^{187}Re^{74+}$, $^{203}Tl^{80+}$, and $^{205}Tl^{80+}$ have also shown big disagreements with theory [7–10]. In order to gain acceptable agreement with theory it was suggested that the value of the nuclear magnetic moment was in need of revision or that the radii of the nuclear magnetization differed by large amounts from those of the nuclear charge. The latter is difficult to comprehend in the view of nuclear models and the typically much smaller assumed differences between the distributions of charged and neutral nucleons. The possibility that at least part of the discrepancies is due to an incorrect understanding of QED could, therefore, not be ruled out.

In order to eliminate the errors of estimating the nuclear magnetization (the so-called Bohr-Weisskopf effect [11]) or the experimental uncertainties in the measured values of the nuclear magnetic moments from obscuring the contributions from QED, Shabaev *et al.* [12] suggested a measurement of the $2s$ hyperfine splitting of the associated lithiumlike ion. They showed that combining the measured energy values of the respective $1s$ and $2s$ ground states cancels out the uncertainties of nuclear parameters, making an unobscured test of the QED effects possible. The theoretical basis for this scheme was further refined recently by Volotka *et al.* [13]. So far, however, no measurement of the $2s$ hyperfine splitting with sufficient accuracy exists to make use of the scheme developed by Shabaev, Volotka, and co-workers. The only successful measurement is that of $^{209}Bi^{80+}$ [14], which determined a value of $0.820 \pm 0.026$ eV for the $2s$ hyperfine splitting using x-ray emission spectroscopy. Subsequent attempts have not yet improved upon the earlier uncertainties [15,16].

In the following, we demonstrate the possibility of utilizing high-resolution spectroscopy of the $2s_{1/2} - 2p_{1/2}$ transition in the extreme ultraviolet to measure the $2s$ hyperfine splitting with a 0.001 eV accuracy. This is, in principle, sufficient to test the accuracy of QED calculations at a level better than a few percent when applying the scheme developed by Shabaev *et al.*, i.e., at a level more than 20 times better than enabled before. Our technique also produces values for the splitting of the $2p_{1/2}$ excited level in lithiumlike ions, which has so far not been measured by any technique. Moreover, it allows us to make the first measurement of the hyperfine splitting of the $2s2p$ $^3P_1$ level in a high-$Z$ berylliumlike ion. Our technique, thus, provides additional experimental information to eliminate the uncertainty from nuclear effects beyond that provided by the $2s$ level in lithiumlike ions.

In order to test our method we have applied it to $^{141}$Pr. $^{141}$Pr has a nuclear spin $I = 5/2$ and, with 82 neutrons, it has a closed neutron shell. However, its proton number ($Z = 59$) is sufficiently low so that the Bohr-Weisskopf and QED effects are predicted to be equal to or smaller than 1 meV. As a result, a comparison between experiment and theory is not obscured by the uncertainty in either and provides an excellent test of the experimental accuracy. An

 

observation of the $F = 2$–3 transition in the $2s$ ground state of the lithiumlike ion would require resonance measurements near 5 $\mu$m, which is not possible to accomplish with laser fluorescence on present-day storage rings. The same is (even more) true for an observation of the $2p_{1/2}$ splitting.

The present experiment was performed at the SuperEBIT high-energy electron beam ion trap at the Lawrence Livermore National Laboratory [17]. The measurements were performed at electron beam energies of about 102 keV and at currents up to about 155 mA. Injection of Pr into the ion trap was achieved by ablation using a pulsed Nd:YAG laser [18]. Praseodymium ions were trapped for about 77 s.

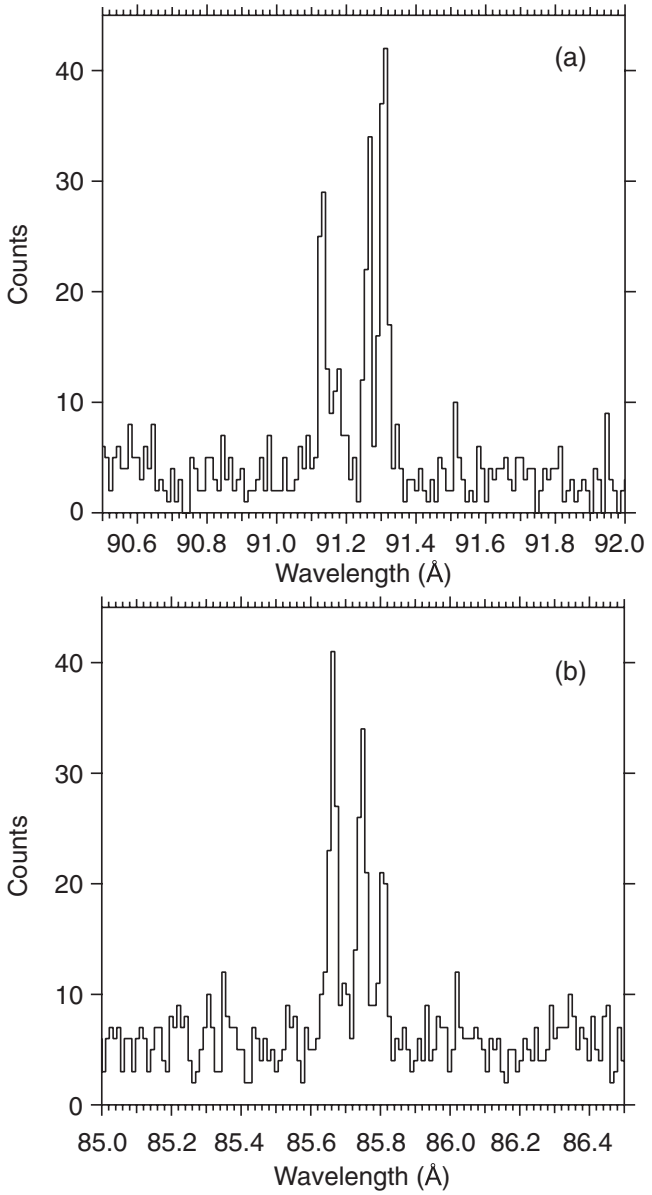


FIG. 1. Spectrum showing 14 h of co-added data: (a) four lines corresponding to the split $2s_{1/2} - 2p_{1/2}$ transition array of $^{141}Pr^{56+}$; (b) three lines corresponding to the $2s^2 {}^1S_0 - 2s2p^3P_1$ transition array of $^{141}Pr^{55+}$.

Then they were dumped, and a new trapping cycle was started. The praseodymium ionization balance was monitored using an x-ray microcalorimeter [19]. Light-ion, evaporative cooling of the praseodymium ions was accomplished by the injection of trimethylborate or nitrogen.

The $2s$ hyperfine splitting in $Pr^{56+}$ amounts to about $1/500$ of the roughly 130 eV $2s_{1/2} - 2p_{1/2}$ transition energy. For our measurements, we employed a high-resolution grazing-incidence spectrometer that utilizes a gold-coated, $R = 44.3$ m grating with a variable line spacing centered around 2400 $\ell$/mm and reflecting at about 2.5°. The instrument was similar to the one described in Ref. [20], except that it was modified to provide better focusing by optimizing the distance between the grating and the SuperEBIT electron beam, which acts as the entrance slit for the otherwise slitless instrument, and to extend its wavelength range well above the previous upper limit of about 50 Å [21]. The spectrometer utilized a cryogenically cooled, back-illuminated charge-coupled device camera with $1340 \times 1300$ 20 $\mu$m wide pixels.

A typical spectrum of the observed $n = 2 - n = 2$ line emission from lithiumlike and berylliumlike praseodymium is shown in Fig. 1. The line width is below 25 mÅ, corresponding to a resolving power above 3600 for the $Pr^{56+}$ lines. The resolving power was slightly worse for the $Pr^{55+}$ lines. The spectrum represents 14 h of co-added data recorded during 1 run day. Each CCD image was typically acquired for 30 or 60 min, then filtered to extract the position of a given photon signal. Our filtering algorithm was specifically constructed to strictly avoid any counting of cosmic ray events. The spectrum clearly shows four $Pr^{56+}$ lines, arising from the splitting of both the $2s$ level and the $2p_{1/2}$ level, and three $Pr^{55+}$ lines, arising from the splitting of the $2s_{1/2}2p_{1/2}{}^3P_1$ upper level. (The $Pr^{55+}$ $2s^2 {}^1S_0$ level is not split, as illustrated in Fig. 2.)

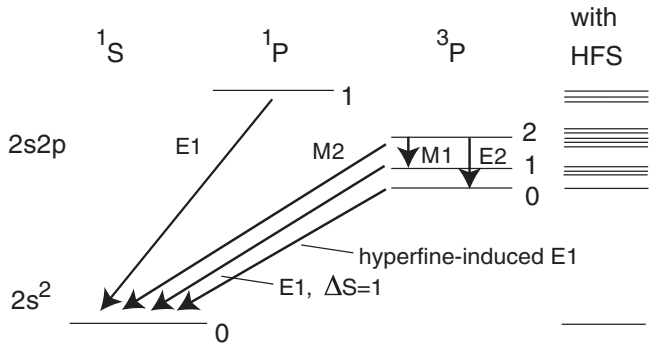


FIG. 2. Schematic of the five lowest levels in berylliumlike $Pr^{55+}$ and their splittings (far right) due to the hyperfine interaction with the nuclear magnetic field. The transition between the $2s^2$ ground state and the $2s2p^3P_1$ upper level splits into three components. Transitions are denoted by their multipole order, where $E1$ and $E2$ ($M1$ and $M2$) denote electric (magnetic) dipole and quadrupole transitions, respectively.

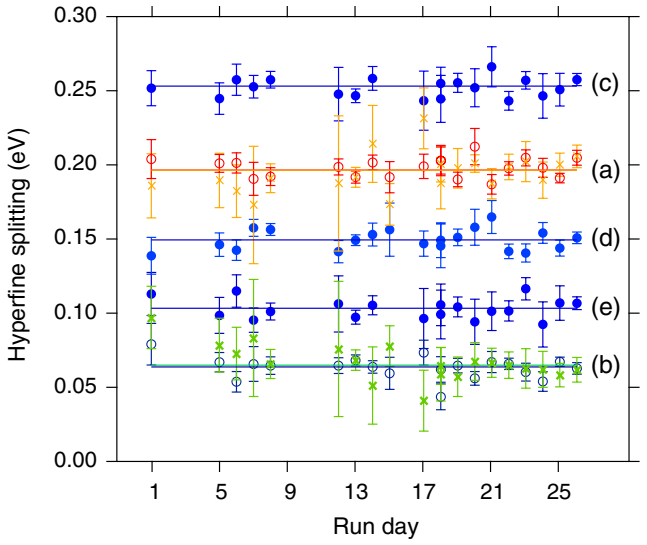


FIG. 3 (color online). Hyperfine splittings derived from data sets collected on different days: (a) $^{141}Pr^{56+}$ hyperfine splitting of the $2s_{1/2}$ ground level (yellow crosses and open orange circles) from the $2s_{1/2} - 2p_{1/2}$ line separations between $F = 2$–2 and 3–2 lines as well as between $F = 2$–3 and 3-3 lines, and (b) of the $2p_{1/2}$ excited level (green crosses and open blue circles) between $F = 2$–2 and 2–3 as well as between $F = 3$–2 and 3–3; (c)–(e) $^{141}Pr^{55+}$ hyperfine splittings of the $2s2p^3P_1$ excited level (solid blue circles) between $F = 3/2$–5/2, 5/2–7/2, and 3/2–7/2. The solid horizontal lines represent the weighted averages for each measured splitting.

The spectrometer wavelength scale and dispersion were derived from the spectral lines of neon produced by injecting the noble gas into the trap under identical operating conditions except that the cycle time was shortened by a factor of 100. The calibration spectra provided seven useful reference lines from lithiumlike, berylliumlike, and boronlike neon, albeit only the lithiumlike lines appear to be known with an accuracy of 1 mÅ or better. In addition, we recorded calibration spectra using a trimethylborate injection, which provided the well known Lyman-$\alpha$ line of hydrogenlike boron as a reference in second order. Useful praseodymium spectra were collected on 20 run days, while calibration data were collected only on 6. Shifts in the spectrometer position from day to day have precluded an accurate absolute wavelength calibration of the praseodymium data. However, in order to determine the hyperfine splitting, it is only necessary to firmly establish the dispersion, which is much more resilient to shifts. Indeed, changes in the dispersion calibration contributed less than 0.2 meV to our measurement uncertainties of a given hyperfine splitting.

The spectral measurements allow us to determine the four spacings between the four $Pr^{56+}$ lines, two of which correspond to the $2s_{1/2}$ and two to the $2p_{1/2}$ splitting, as well as the three line spacings for the three $Pr^{55+}$ lines. The seven hyperfine splittings inferred for each of the 20 useful run days are presented in Fig. 3 together with the weighted average for each of the 7 splittings. The numerical values of the weighted averages for the lithiumlike and berylliumlike splittings are given in Tables I and II, respectively.

The experimental uncertainties are essentially all from counting statistics, which is also reflected in the error bars shown in Fig. 3. The aforementioned error in the dispersion was determined by recalculating all splittings for the six available calibration curves and then adding the differences in quadrature.

Tables I and II also list the theoretical values for each hyperfine splitting we have calculated using the relativistic configuration-interaction (RCI) method [22]. To gauge the accuracy of our RCI results, we first note that we use a nuclear magnetic moment of $4.2754 \pm 0.0005$ n.m. from the tabulation of Raghavan [23], which should be accurate enough as to not cause any significant theoretical uncertainties. $^{141}Pr$ also has an electric quadrupole moment of $-0.0589 \pm 0.0042$ barn [23], but it has no effect on the $J = 1/2$ states and the $E2$ contribution to the hyperfine splitting of the $^3P_1$ state is less than 0.1 meV and can be ignored. As for the electron correlation effects, they are not expected to be very important for high-$Z$ Li- and Be-like ions and should be accurately accounted for by the present highly correlated RCI calculations to within 0.1 meV. The most uncertain contributions are those from the Bohr-Weisskopf [11] and QED effects. Here, we assume that the nuclear magnetization radius is the same as the nuclear charge radius, which we take to be 6.3154 fm based on the root-mean-square nuclear radius of $4.8919 \pm 0.0050$ fm from the tabulation of Angeli [24]. Resulting

TABLE I. Predicted and measured values (in meV) of the hyperfine splitting of the $2s_{1/2}$ and $2p_{1/2}$ levels in Li-like $^{141}Pr^{56+}$.

| Interval | Source | Hyperfine splitting | Uncertainty | Reference |
|---|---|---|---|---|
| $2s_{1/2}$ $F = 2$–3 | Experiment | 196.5 | 1.2 | This work |
| | Theory | 197.4 | 0.5 | [25] |
| | Theory | 197.5 | | [26] |
| | Theory | 197.3 | 1.0 | This work |
| $2p_{1/2}$ $F = 2$–3 | Experiment | 64.0 | 1.1 | This work |
| | Theory | 63.62 | 0.07 | [27] |
| | Theory | 63.6 | 0.2 | This work |

TABLE II. Predicted and measured values (in meV) of the hyperfine splitting of the $2s_{1/2}2p_{1/2}\,^3P_1$ level in Be-like $^{141}Pr^{55+}$. All results are from this work.

| Interval | Source | Hyperfine splitting | Uncertainty |
|---|---|---|---|
| $F = 5/2$–$7/2$ | Experiment | 149.4 | 1.4 |
| | Theory | 148.7 | 1.0 |
| $F = 3/2$–$5/2$ | Experiment | 103.3 | 1.9 |
| | Theory | 106.2 | 1.0 |
| $F = 3/2$–$7/2$ | Experiment | 253.1 | 1.8 |
| | Theory | 254.9 | 1.0 |

Bohr-Weisskopf corrections are found to be small at about 1 meV, specifically, 1.6 and 0.3 meV for the Li-like $2s_{1/2}$ and $2p_{1/2}$ hyperfine splittings, respectively, and 0.7 and 0.5 meV for the Be-like $F = 5/2 - 7/2$ and $3/2 - 5/2$ splittings, respectively. QED corrections are likewise small at 0.9 meV for the Li-like $2s$ splitting and 0.5 and 0.4 meV for the same Be-like splittings mentioned earlier. The QED correction for the $2p_{1/2}$ splitting is quite negligible at less than 0.1 meV. At this level, the errors inherent in the calculation of these two corrections should have no effect on the accuracy of the predicted hyperfine splittings. In the following, we conservatively assume an error about half the size of these effects, i.e., 1 meV for the Li-like $2s_{1/2}$ and Be-like $^3P_1$ hyperfine splittings, and 0.2 meV for the Li-like $2p_{1/2}$ splittings. For the $2s_{1/2}$ hyperfine interval in lithiumlike $Pr^{56+}$, our calculations agree very well with the calculated values from Shabaev *et al.* [25] and from Boucard and Indelicato [26]. Similarly, our calculations agree very well with the prediction by Korzinin *et al.* [27] for the $2p_{1/2}$ hyperfine splitting. Their uncertainties are less conservatively estimated at 0.07 meV. We are not aware of earlier calculations for the hyperfine splitting in berylliumlike $Pr^{56+}$.

Comparing the measured values with the predicted values, we find excellent agreement. Four of the measured values agree well within the experimental uncertainty. For example, the measured $2s$ hyperfine splitting of $196.5 \pm 1.2$ meV compares well with our prediction of $197.3 \pm 1.0$ meV, and the measured value of the $2p_{1/2}$ splitting of $64.0 \pm 1.1$ meV compares well with the predicted value of $63.6 \pm 0.2$ meV. The one exception is the measured value for the $F = 3/2 - 5/2$ splitting in berylliumlike praseodymium, which we measured to be $103.3 \pm 1.9$ meV, i.e., 2.9 meV less than our calculated value $106.2 \pm 1.0$ meV, but still within the range expected for 1-$\sigma$ measurements.

In summary, we have demonstrated that EUV spectroscopy can be used to measure the hyperfine structure in high-$Z$, few-electrons ions at the meV level. The excellent agreement with theory not only validates the technique but also shows that in the near absence of QED and Bohr-Weisskopf effects theory can correctly treat the hyperfine splitting.

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*beiersdorfer1@llnl.gov
†Also at Ruhr-Universität Bochum, D-44780 Bochum, Germany.

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