Measurements of $2s \, {}^{2}S_{1/2} - 2p \, {}^{2}P_{1/2,3/2}$ transition energies in lithiumlike heavy ions. II. Experimental results for Ag⁴⁴⁺ and discussion along the isoelectronic series

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Wavelengths of the fine structure transitions $2s^2S_{1/2}$ - $2p^2P_{3/2}$ and $2s^2S_{1/2}$ - $2p^2P_{1/2}$ in lithiumlike Ag⁴⁴⁺ have been measured using beam-foil excitation and grazing incidence spectroscopy. The respective transition wavelengths of 40.829 ± 0.004 Å and of 124.685 ± 0.009 Å were not measured before for this ion. The achieved precision of 70 ppm for the 1/2-1/2 transition allows for a 0.2% QED test which sets a new benchmark in the medium *Z* range. Our experimental uncertainty is smaller than calculated two-photon exchange contributions to the transition energy. A summary of all published experimental data along the isoelectronic sequence for $Z \ge 24$, including our recent results for Ni²⁵⁺ and Zn²⁷⁺, is presented and compared with the best available calculations. [S1050-2947(99)02703-1]

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I. INTRODUCTION

The study of the two fine-structure transitions $2s^{2}S_{1/2}-2p^{2}P_{1/2}$ (1/2-1/2) and $2s^{2}S_{1/2}-2p^{2}P_{3/2}$ (1/2-3/2) of lithiumlike ions is of high interest for testing atomic structure theories. Abundant accurate experimental spectroscopic measurements for lithiumlike ions with atomic numbers Z <36 are available. The most precise results with a relative accuracy of 30 ppm were obtained for Z=24, 26, and 28 using grazing incidence spectroscopy at tokamak generated plasmas [1-3]. These experiments are sensitive to 0.3% of the quantum electrodynamic (QED) contribution to the transition energy of the 1/2-1/2 transition. For the highest Z values, accurate data are available only for Z=83, 90, and 92 for the 1/2-3/2 transition resulting from x-ray spectroscopy of ions produced at an electron beam ion trap (EBIT) [4-6]. For the 1/2-1/2 transition, a single result for uranium has been reported utilizing Doppler tuned spectroscopy of fast ions [7]. This measurement gives a QED sensitivity of 0.2%. In the range of intermediate atomic numbers (36 < Z < 83)only one significant measurement exists at Z=42 [8]. The only two other experiments in the medium Z range were performed with Xe ions using beam foil spectroscopy [9,10], but they suffer from large error bars and provide only poor OED sensitivity. We have recently reported accurate beam foil measurements of 2s-2p transitions in lithiumlike Ni²⁵⁺ and Zn^{27+} (paper I) [11]. The present paper presents the only measurement so far, to our knowledge, of the 2s-2p finestructure transition energies in Ag44+. Our results provide the highest QED sensitivity for intermediate lithiumlike ions.

Below we will describe the measurement procedure and

report the results for the fine-structure transitions in lithiumlike Ag⁴⁴⁺. Then a compilation of all published experimental transition energies for $Z \ge 24$ is given for the lithiumlike isoelectronic sequence. It includes our results for Ag⁴⁴⁺ as well as our recent values for Ni²⁵⁺ and Zn²⁷⁺ published in paper I [11]. All these experimental data are then compared to calculations by Chen *et al.* [12], Blundell [13], and Kim *et al.* [14].

II. EXPERIMENT

We report only briefly on the experimental setup and the measurement procedure. For details we refer to paper I [11]. The experiment was carried out at the heavy ion accelerator UNILAC at GSI with 13.247 MeV/nucleon Ag¹⁵⁺ ions impinging on a 415 μ g/cm² carbon foil. The vacuum ultraviolet (vuv) light emitted from the excited highly charged silver ions was analyzed using the GSI 5 m grazing incidence spectrometer. We use a Rowland circle geometry with a 270 lines/mm spherical grating to disperse the light onto a movable two-dimensional microchannel-plate detector. A Penning discharge lamp, which is periodically blocked by a chopper wheel synchronized with the ion beam pulse, serves as an *in situ* wavelength scale calibration standard. It is operated with a He-Ne gas mixture and Al cathodes to produce accurately known intense calibration lines [15–17].

Figure 1 shows a spectrum in the vicinity of the 1/2-1/2 transition in the second order of diffraction, recorded within 4 h. A calibration spectrum recorded in first order, synchronously and simultaneously with the projectile spectrum, is also shown. A total of 17 runs was carried out for this transition. The line to the left of the assigned transition is not yet

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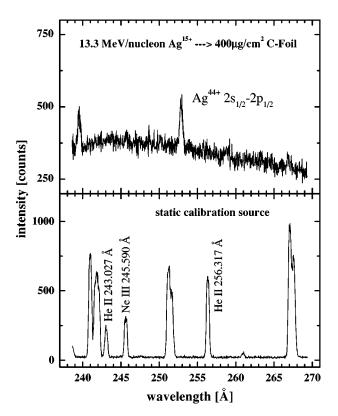


FIG. 1. Beam foil spectrum of the $2s \, {}^{2}S_{1/2} - 2p \, {}^{2}P_{1/2}$ transition of lithiumlike Ag⁴⁴⁺ accumulated for 4 h together with a simultaneous calibration spectrum. The assigned He II and Ne III lines from the static plasma of the Penning source were used to calibrate the Doppler shifted projectile line observed in second order of diffraction.

identified. Figure 2 shows one of seven 1/2-3/2 transition spectra each recorded within 1 h. This line was observed in the fourth order. Also depicted is the corresponding calibration spectrum in first order. The average ion current was about 2×10^{14} ions/s.

A precise wavelength scale was established by a quadratic regression using three calibration lines. For the 1/2-1/2 transition the 245.590(1) Å Ne III line [15,16] and the two He II lines at 243.027(1) Å and 256.317(1) Å [17,18] were utilized. For the 1/2-3/2 transition the Al III line at 169.070(10) Å and two Al IV lines at 160.074(4) Å and 161.688(4) Å [19] were used. The calibration uncertainty was determined by systematically shifting the centroid of the calibration lines within their statistical and wavelength uncertainties and taking the standard deviation of the resulting lithiumlike transition wavelength. An uncertainty of 0.007 Å was determined for the 1/2-1/2 transition and 0.008 Å for the 1/2-3/2 transition.

In our previous experiments it was necessary to correct the spectra for a deviation of the detector response from linearity (see paper I). By using a new wedge and strip anode and a refined adjustment technique for the detector, we were able to reduce the nonlinearity below the statistical error of the line position.

Due to the high velocity of the ions (β =0.17) the projectile lines appear Doppler shifted. To correct for this shift, the angle of observation as well as the projectile velocity must be known accurately. The angle of observation is therefore

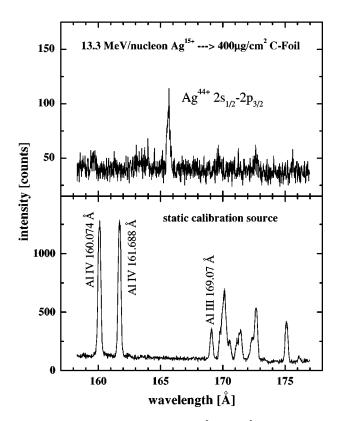


FIG. 2. Beam foil spectrum of the $2s {}^{2}S_{1/2} - 2p {}^{2}P_{3/2}$ transition of lithiumlike Ag⁴⁴⁺ accumulated for 1 h together with a simultaneous calibration spectrum. The assigned Al III and Al IV lines from the static plasma of the Penning source were used to calibrate the Doppler shifted projectile line observed in fourth order of diffraction.

carefully aligned to $(90.00\pm0.01)^\circ$ with respect to the ion beam axis by an elaborate optical adjustment procedure described in paper I. The actual direction of the ion beam as well as the velocity of the projectiles are measured during each of the 24 runs. The projectile velocity is measured applying a time-of-flight method with two pickup coils separated by 13 m.

We performed a series of additional runs dedicated to checking the spectrometer performance and to confirming that the correct and undisturbed transition lines were observed. Most importantly we made use of the possibility to turn the spectrometer around the optical axis by 180°. Thus we were able to consider shifts of the Doppler broadened projectile lines with respect to the static calibration lines. These shifts can occur due to lifetime effects or if the efficiency of the grating varies locally. A negligible grating effect has previously been observed at 160 Å and at 230 Å [20]. The lifetime of the 1/2-1/2 transition at 125 Å amounts to 240 ps. This is long with respect to the observed decay length of 2 ps. Consequently, for the two grating orientations, no wavelength shift outside the statistical errors was observed for the 1/2-1/2 transition. The situation is different for the 1/2-3/2 transition at 41 Å. Here we did observe a wavelength difference of 0.02 Å between the two rotation positions, which can be partly attributed to the short lifetime of the ${}^{2}P_{3/2}$ state of 8 ps [21]: The intensity variation due to the lifetime amounts to 25% over the observed decay length,

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TABLE I. Experimental results for the wavelengths of the $2s^2S_{1/2}$ - $2p^2P_{1/2,3/2}$ transitions in lithiumlike Ag⁴⁴⁺ ions (in angstroms) compared with calculations.

Ag ⁴⁴⁺	1/2-1/2	1/2-3/2
This work	124.685(9)	40.829(4)
Theoretical		
Kim et al. (Ref. [14])	124.698	40.832
Blundell ^a (Refs. [13,32])	124.670	40.826
Chen ^b (Refs. [12,30])	124.695	40.829

^aInterpolation carried out by Blundell.

^bInterpolation carried out by Cheng.

and the resulting wavelength difference of 0.01 Å explains half of the shift. But little is known about the local grating efficiency at 41 Å. An assumed variation of the grating reflectivity of 25% could explain the remaining 0.01 Å shift. The possibility of turning the spectrometer around the optical axis allows us to average the values obtained for the two different spectrometer positions, making it unnecessary to model the grating behavior in detail. Therefore, we regard this turning and averaging procedure as crucial for a highprecision measurement.

III. RESULTS FOR Ag44+

The spectra were fitted, calibrated, and Doppler corrected for each run separately. The result for the 1/2-1/2 transition was obtained by determination of the weighted average over all 17 runs. The weight of each individual run contains mainly the statistical uncertainty, but also contributions of beam position and energy uncertainty. The result for the seven runs of the 1/2-3/2 transition was obtained in a different way. First the weighted average was calculated for the two turning positions separately, then the final transition wavelength was determined by averaging the two values. Results for the 2s-2p fine-structure transitions are given in Table I together with theoretical predictions [12–14]. For the 1/2-3/2 transition, agreement within one experimental standard deviation is found for all three calculations. For the 1/2-1/2 transition our experimental value lies halfway between the calculations of Kim *et al.* and Chen *et al.* on the one side and Blundell on the other side. Agreement with all three calculations is only within two experimental standard deviations. The different contributions to the experimental uncertainty are given in Table II. For a detailed discussion of these uncertainties we refer to paper I [11].

IV. THE LITHIUMLIKE SEQUENCE: COMPARISON OF EXPERIMENTAL RESULTS WITH THEORY

In the following we discuss the present status of 2s-2ptransition experiments of lithiumlike heavy ions for $Z \ge 24$ and their significance for testing atomic structure theory. The total transition energies of 2s-2p transitions are considerably smaller than the 1s-2p transitions in hydrogenlike ions thereby increasing the relative amount of QED contributions (e.g., in lithiumlike U^{89+} the relative 2s electron QED contribution is as large as 15% whereas the relative 1s electron Lamb shift in hydrogenlike U^{91+} is only 0.3%). In the theoretical description of hydrogenlike ions, relativistic and QED contributions as well as nuclear effects have to be evaluated. Since lithiumlike ions are three-electron systems, additional terms must be considered. These are electron correlation terms and QED screening effects involving an additional photon exchange between valence and core electrons. The additional energy corrections are of the order α^2 . At intermediate Z they amount to 10% of the first-order terms-the one electron self-energy and vacuum polarization.

At present, the experimental uncertainties of experiments are of the order of the differences of the different theories. Therefore, a careful study of the transition energies along the isoelectronic sequence is needed to increase the significance of all experiments together by supplying precise experimental values for many different Z values. That way not only irregularities in the experimental data can be identified, but also the Z scaling of as yet uncalculated terms can be estimated.

The following discussion of experimental results is carried out in three steps for each transition separately. First the predictions of the different relative contributions to the total

TABLE II. Summary of individual contributions to the experimental uncertainties of the 2s-2p finestructure transition wavelengths in lithiumlike Ag^{44+} . The ${}^{2}S_{1/2} {}^{-2}P_{1/2}$ transition was measured in second order and the ${}^{2}S_{1/2} {}^{-2}P_{3/2}$ transition in fourth order of diffraction.

Error source	Contributions $\times (10^{-3} \text{ Å})$	
Ag^{44+}	1/2-1/2	1/2-3/2
Statistics in beam spectrum	6	9
Statistics in calibration spectrum	7	3
Uncertainty of calibration line wavelengths	2	7
Total for Doppler shifted wavelength	9	12
Adjustment of the angle of observation (0.26 mrad)	11	7
Reproducibility of the angle of observation (0.05 mrad)	2	1
Determination and stability of the center of the ion beam (0.10 mrad)	4	3
Ion beam velocity behind the excitation foil	9	6
Total for Doppler correction	15	10
Total	17	15

transition energy are given for the two most advanced calculations of Blundell [13] and Chen *et al.* [12] along the Liisoelectronic series. The relative uncertainties of experiments are included in the figures to demonstrate the sensitivity of the measurements to the magnitudes of calculated terms. Then the differences between experiment and theory are shown normalized to the QED contribution. Finally QED and non-QED contributions (as grouped together by the authors of the three calculations) are compared for the different calculations in order to decide whether the non-QED contribution is established accurately enough to identify an "experimental QED contribution." All 2s-2p experimental data that are used in the discussion are summarized in Table III.

Before we continue the discussion of the lithiumlike series, we comment briefly on the situation for heliumlike 2s-2p transitions. Two studies which compare experimental and theoretical results were carried out along the isoelectronic sequence. Berry et al. [22] revealed a systematic difference between experiment and the results of the most advanced calculation up to then by Drake [23] for the $1s2s {}^{3}S_{1}-1s2p {}^{3}P_{0}$ transition. With respect to QED sensitivity, this transition resembles the 1/2-1/2 transition of the lithiumlike sequence. The experimental result for Z=47 by Marrus *et al.* [24] has a relative uncertainty which is a factor of 10 larger than our result for the lithiumlike 1/2-1/2 transition with the QED contribution being of comparable magnitude. Kukla et al. [25] were recently summarizing experiments at $3 \le Z \le 36$ and were comparing them with the calculations of Plante et al. [26]. In their measurement of the $1s2s^{3}S_{1}-1s2p^{3}P_{0}$ transition in heliumlike argon they achieved a QED sensitivity of better than 0.5%.

A. The 1/2-1/2 transition

The most successful methods to calculate transition energies for lithiumlike ions so far have been the multiconfiguration Dirac-Fock (MCDF) method applied by Kim et al. [14], the relativistic many body perturbation theory (RMBPT) applied by Blundell [13], and a relativistic configuration interaction (CI) calculation by Chen et al. [12]. Kim et al. calculated transition energies for all Z, whereas Blundell and Chen et al. performed their calculations only for selected Z values. Several publications have concentrated on Z=92, where in a recent paper by Persson *et al.* some two-loop contributions are included [27]. The QED contributions calculated by Kim *et al.* are primarily based on a scaling of Mohr's hydrogenic self-energies [28]. Blundell and Chen et al. evaluated their QED contributions using ab initio calculations of the screening corrections. In the following comparisons we will not use interpolations (unless explicitly stated and performed by the authors of the original papers), since we have observed irregularities in our own interpolations.

The different contributions to the transition energy of lithiumlike ions for the 1/2-1/2 transition are plotted in Fig. 3 for the CI calculation of Chen *et al.* and in Fig. 4 for the RMBPT calculation of Blundell. Chen *et al.* list four different contributions to the total transition energy: The dominant term is the Coulomb interaction containing a no-pair Hamiltonian with a Fermi charge distribution as nuclear potential. The two second strongest terms represent the total QED con-

tribution and the Breit interaction. Both are of almost equal magnitude for all Z values. The smallest contribution is the mass polarization (MP) term. The QED contribution increases roughly as Z^4 from 1% at Z=25 to 15% at Z=92. The Breit term, which describes electron-electron interactions, contains frequency-dependent retarded Breit energies of higher orders, which are not included in the other two calculations.

Figure 4 shows the relative contributions to the total 1/2-1/2 transition energy included in the calculation of Blundell [13]. The dominant part in the calculation is the RMBPT term, which includes also some higher-order Breitinteraction terms. The QED contribution is dominated by the valence self-energy (SE) term. Contributions from twophoton exchange Feynman diagrams are of the order of 0.1%-2% of the QED contribution. In these terms one additional photon is exchanged between core and valence electrons. The terms labeled "X," "C," and "Other" are Blundell's notation for subsets of all screening correction terms of the order of α^2 , but there remain uncalculated terms (for details refer to Ref. [13]).

The relative accuracies of all experiments with $Z \ge 24$ known to us are also depicted in Figs. 3 and 4, in order to compare them with the magnitudes of the calculated contributions. Different symbols denote different experimental techniques. A detailed discussion of lower Z data is given in Ref. [29]. Spectroscopic measurements on solar flares, with uncertainties greater than 100 ppm, were the first measurements above Z=20 [29]. In the region of $24 \le Z \le 36$, accurate tokamak data [1-3,8,35] as well as our beam foil measurements for Z=28 and 30 provide a sound basis to test theoretical predictions. Above Z=36 only three significant measurements for Z=42 [8] and Z=47 and Z=92 [7] exist with relative accuracies of 140 ppm, 70 ppm, and 360 ppm, respectively. The molybdenum measurement is the highest Zresult from a tokamak plasma, since the plasma temperatures do not allow an efficient production of heavier lithiumlike ions. Our new beam foil measurement on Ag ions has the smallest relative uncertainty to date in medium Z 1/2-1/2measurements. The measurement in lithiumlike uranium was performed with the Doppler tuned technique. In the medium Z range, two other beam foil experiments with Xe exist [9,10], but their experimental uncertainty is an order of magnitude larger than for the most precise measurements. We note that the screening-correction effects shown in Fig. 3 are larger than the uncertainties of the best transition energy measurements, including this experiment for Z=47.

It is evident that without any further assumption, the experimental transition energies can only be compared to the calculated total transition energies. This comparison is shown in Fig. 5 for the 1/2-1/2 transition and the three discussed calculations [12–14]. In contrast to Figs. 3 and 4, we considered only experiments with a precision of better than 2% of the QED contribution. We normalized the difference in transition energies $\Delta = E^{\text{expt}} - E^{\text{theor}}$ to the absolute value of the QED energy contribution $|E_{\text{QED}}^{\text{theor}}|$. The experimental error bars are also scaled this way. As a consequence, this comparison of experiments for different Z leads to a reduction of the size of the error bars of the experiment with increasing Z.

TABLE III. Summary of experimental 2s-2p fine-structure transition energies for lithiumlike ions (in cm⁻¹).

	1/2-1/2 1/2-3/2				
Ζ	Energy	Uncertainty	Energy	Uncertainty	Author
24	357525	38	448461	60	Dere et al. (Ref. [29])
	357537	38	448441	60	Widing et al. (Ref. [29])
	357537	38	448461	60	Sandlin et al. (Ref. [29])
	357539	26	448430	40	Davé et al. (Ref. [35])
	357489	25	448410	40	Hinnov et al. (Ref. [8])
			448392	12	Knize et al. (Ref. [1])
	357484	6	448396	10	Sugar et al. (Ref. [2])
25	374734	42	483393	70	Dere et al. (Ref. [29])
	374706	42	483323	70	Widing et al. (Ref. [29])
	374706	42	483346	70	Sandlin et al. (Ref. [29])
	374700	50	483320	50	Sugar <i>et al.</i> (Ref. [14])
26	391988	46	520760	81	Dere et al. (Ref. [29])
	391988	46	520733	81	Widing et al. (Ref. [29])
	392003	46	520760	81	Sandlin et al. (Ref. [29])
	392049	31	520806	54	Davé et al. (Ref. [35])
	392012	15	520800	55	Hinnov et al. (Ref. [8])
	391986	15	520708	24	Knize et al. (Ref. [1])
	391983	8	520757	14	Reader et al. (Ref. [3])
28	427150	109			Zacharias et al. (Ref. [36])
	427068	18	604610	36	Hinnov et al. (Ref. [8])
	427071	9	604595	18	Sugar et al. (Ref. [2])
			604928	80	Büttner et al. (Ref. [37])
	427044	31	604573	55	Staude et al. (Ref. [11])
29	444850	20	651436	85	Hinnov et al. (Ref. [8])
	444891	22	651440	25	Knize et al. (Ref. [1])
30	462832	24	701946	30	Staude et al. (Ref. [11])
32			814744	100	Behring et al. (Ref. [38])
	499276	25	814963	130	Hinnov et al. (Ref. [8])
	499264	15	814963	40	Knize et al. (Ref. [1])
34	536552	45	946199	180	Hinnov et al. (Ref. [8])
	536596	20			Knize et al. (Ref. [1])
36	574218	857	1097936	1205	Dietrich et al. (Ref. [39])
	574594	85	1098310	300	Hinnov et al. (Ref. [8])
	574944	132	1098901	362	Martin et al. (Ref. [40])
			1099022	966	Büttner et al. (Ref. [41])
			1098479	833	Büttner et al. (Ref. [37])
42	694454	96	1709431	585	Hinnov et al. (Ref. [8])
47	802021	58	2449240	240	this work
54	967586	749	3971406	4732	Martin et al. (Ref. [9])
	967118	1871	3969829	7880	Büttner et al. (Ref. [10])
83			22487849	315	Beiersdorfer et al. (Ref. [6])
90			32465657	1129	Beiersdorfer et al. (Ref. [5])
92	2263110	807			Schweppe et al. (Ref. [7])
			35967231	1694	Beiersdorfer <i>et al.</i> (Ref. [4])

The basic idea of normalizing Δ to $|E_{QED}^{\text{theor}}|$ is to show the QED sensitivity of experiments, since Δ can be written as $\Delta = E^{\text{expt}} - E_{\text{nonQED}}^{\text{theor}} - E_{QED}^{\text{theor}}$. Assuming that the non-QED part of the transition energy $E_{\text{nonQED}}^{\text{theor}}$ can be calculated with

much higher precision than the QED terms $E_{\text{QED}}^{\text{theor}}$ (we discuss this in Fig. 6 below), one can identify an experimental QED contribution $E_{\text{QED}}^{\text{expt}} = E^{\text{expt}} - E_{\text{nonQED}}^{\text{theor}}$. Then Δ can be interpreted as the difference between experimental and calculated

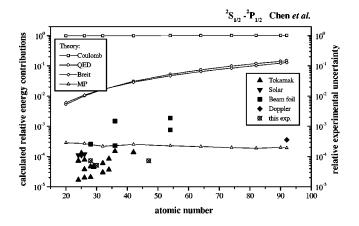


FIG. 3. Comparison of relative contributions to the total $2s_{1/2}^{-2}P_{1/2}$ transition energy as a function of *Z* calculated by Chen *et al.* (Ref. [12]). The relative uncertainties of all available experiments are included, where different symbols denote different spectroscopic techniques. Tokamak: *Z*=24 (Refs. [35,8,2]), *Z*=25 (Ref. [14]), *Z*=26 (Refs. [35,8,1,3]), *Z*=28 (Refs. [8,2]), *Z*=29 (Refs. [8,1]), *Z*=32 (Refs. [8,1]), *Z*=34 (Refs. [8,1]), *Z*=36 (Ref. [8]), *Z*=42 (Ref. [8]); solar flares: *Z*=24,25,26 (Ref. [29]); beam foil: *Z*=28 (Refs. [36,11]), *Z*=30 (Ref. [11]), *Z*=36 (Refs. [39,40]), *Z*=47 (this work), *Z*=54 (Refs. [9,10]); Doppler tuned: *Z*=92 (Ref. [7]).

QED values, since $\Delta = E_{\text{QED}}^{\text{expt}} - E_{\text{QED}}^{\text{theor}}$. Under the above assumption, experiment can distinguish between different calculations of $E_{\text{OED}}^{\text{theor}}$.

Since Kim *et al.* calculated energies for every single Z value, this theory can be tested with all available highprecision data (12 different Z values). And even though they did not perform *ab initio* calculations of E_{QED}^{theor} , the overall agreement between experiment and the calculations is rather satisfactory. This is especially remarkable because the computing time is more than two orders of magnitude smaller compared to the CI calculations of Chen *et al.* [12]. The calculation of Chen *et al.* shows no systematic deviation from the experiments for the five Z values where measurements exist. For Z=47, no theoretical results are given in the original paper [12] and we used an interpolation by one of

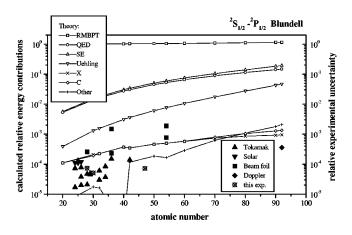


FIG. 4. Comparison of relative contributions to the total ${}^{2}S_{1/2}{}^{-2}P_{1/2}$ transition energy as a function of *Z* calculated by Blundell (Ref. [13]). The relative uncertainty of experiments is included. (For references see the caption of Fig. 3.)

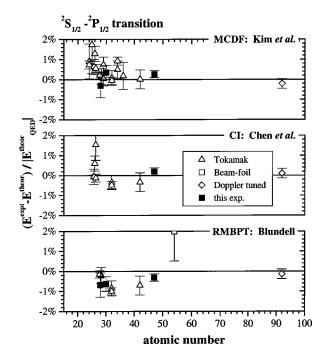


FIG. 5. Normalized difference between experimentally determined and calculated $2s {}^{2}S_{1/2} - 2p {}^{2}P_{1/2}$ transition energies along the Li isoelectronic sequence. Data points are shown only where theoretical and experimental values exist for the same atomic number. (For references of experiments, see the caption of Fig. 3). Calculations are from Kim *et al.* (Ref. [14]), Chen *et al.* (Ref. [12]), and Blundell (Ref. [13]).

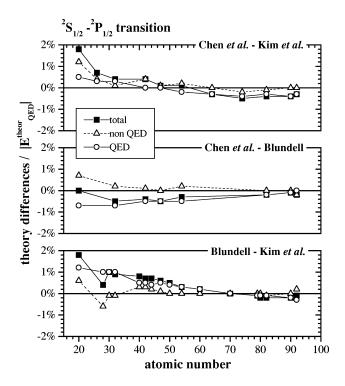


FIG. 6. Normalized differences of the theoretical predictions for the QED contribution, the non-QED part, and the total energy for the $2s^{2}S_{1/2}$ - $2p^{2}P_{1/2}$ transition along the lithiumlike sequence. The calculations were performed by Chen *et al.* (Ref. [12]), Blundell (Ref. [13]), and Kim *et al.* (Ref. [14]).

the authors [30]. Nuclear polarization energies are omitted from the calculated values, since they had been overestimated by a factor of 2π [31] making this contribution insignificant. Blundell's values [13] at intermediate Z tend to be about 0.5% higher than the measured transition energies. However, at Z=92 good agreement is found with the calculation. For Z=28 and 47 we have used values interpolated by Blundell [32].

The remaining question is whether the non-QED contributions to the transition energy are known precisely enough to allow a significant test of the calculated OED terms. Figure 6 shows a comparison of the QED contributions, the non-QED parts, and the sum of both for the three different calculations. The differences of the theoretical predictions are normalized to the absolute value of the QED contribution and plotted along the isoelectronic sequence, wherever two calculations have provided energies for the same Z. Above Z=20, the non-QED parts of the three calculations agree within 0.2%. Only the interpolated value of Blundell at Z=28 shows a greater deviation. Especially from the agreement with the non-OED contributions of Chen et al., it follows that higher-order Breit terms (contained only in the calculation of Chen et al.) do not contribute significantly to the 1/2-1/2 transition energy. Due to the very good agreement, we consider the non-QED part to be known with sufficient accuracy to assign the differences between calculated and experimentally determined 1/2-1/2 transition energies depicted in Fig. 5 primarily to the OED terms. We note that the more systematic QED calculation of Blundell deviates significantly more from experimental results than does the calculation of Chen. Here one could even deduce a 1/Z dependence of $(E^{\text{expt}} - E^{\text{theor}})/E_{\text{OED}}^{\text{theor}}$, which could result from the Z^3 dependence of uncalculated screening correction terms normalized to the Z^4 -dependent QED contributions.

B. The 1/2-3/2 transition

In Figs. 7 and 8 the different contributions to the transition energy of lithiumlike ions for the 1/2-3/2 transition are plotted for the CI calculation by Chen et al. [12] and for the RMBPT calculations by Blundell. The major difference between the 1/2-3/2 transition and the 1/2-1/2 transition in Fig. 3 is that the relative QED contribution remains at the 1% level for all Z>25. Even though the absolute value of the QED contribution still increases with Z^4 as for the 1/2-1/2transition, it is compensated by the fine-structure splitting of the 2p levels which also scales with Z^4 , increasing the total 1/2-3/2 transition energy. As a consequence, a much higher experimental accuracy has to be achieved to reach the OED sensitivity of high-Z measurements of the 1/2-1/2 transition. The Breit contribution given by Chen et al. (Fig. 7) amounts to only 20%-40% of the QED energy. The mass polarization term is for low Z on the 100 ppm level, but becomes insignificant for the highest Z values. Also depicted in Figs. 7 and 8 is the relative accuracy of all experiments known to us, where different symbols again denote different experimental techniques. In Fig. 8 the single QED contributions to the 1/2-3/2 transition energy are shown as calculated by Blundell [13]. A comparison with Fig. 4, where the same contributions are given for the 1/2-1/2 transition, shows that the two-

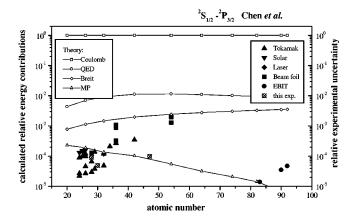


FIG. 7. Comparison of relative contributions to the total ${}^{2}S_{1/2}{}^{-2}P_{3/2}$ transition energy as a function of *Z* calculated by Chen *et al.* (Ref. [12]). The relative uncertainties of all available experiments are included, where different symbols denote different spectroscopic techniques. Tokamak: *Z*=24 (Refs. [35,8,1,2]), *Z*=25 (Ref. [14]), *Z*=26 (Refs. [35,8,1,3]), *Z*=28 (Refs. [8,2]), *Z*=29 (Refs. [8,1]), *Z*=32 (Refs. [8, 1]), *Z*=34 (Ref. [8]), *Z*=36 (Ref. [8]), *Z*=42 (Ref. [8]); solar flares: *Z*=24,25,26 (Ref. [29]); laser plasma: *Z*=32 (Ref. [38]); beam foil: *Z*=28 (Refs. [37,11]), *Z*=30 (Ref. [11]), *Z*=36 (Refs. [39–41,37]), *Z*=47 (this work), *Z*=54 (Refs. [9,10]); EBIT: *Z*=83,90,92 (Refs. [4–6]).

photon exchange terms roughly scale with the total QED contribution and thus remain on the 10^{-4} level.

As in Fig. 5, we show in Fig. 9 the difference between theoretical predictions and the most precise experiments. Again, this difference is normalized to the absolute value of the QED energy contribution and only experiments with uncertainties smaller than 2% in "QED units" are shown. Our measurement for Ag^{44+} is in agreement with all three calculations. The only other experiments at high Z are the EBIT measurements of Beiersdorfer *et al.* for Z=83, 90, and 92 with accuracies of 14 ppm, 35 ppm, and 47 ppm, respectively [4–6]. All the other experiments suffer from the increase of the transition energy and the corresponding larger error bars. The Z=83 data point is clearly not in agreement with the calculation of Kim *et al.* Unfortunately, neither Blundell nor Chen *et al.* have given a value for this ion. In

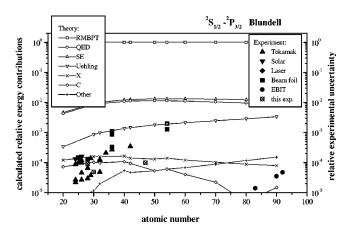


FIG. 8. Comparison of relative contributions to the total ${}^{2}S_{1/2}{}^{-2}P_{3/2}$ transition energy calculated by Blundell (Ref. [13]) as a function of Z. The relative uncertainties of the experiments are included. (For references, see the caption of Fig. 7.)

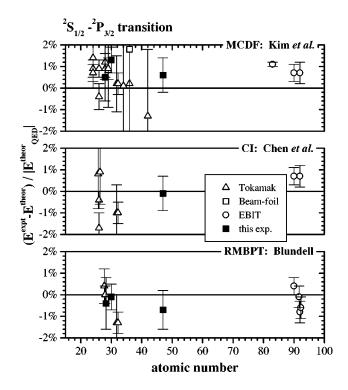


FIG. 9. Normalized difference between experimentally determined and calculated $2s \, {}^{2}S_{1/2} - 2p \, {}^{2}P_{3/2}$ transition energies along the Li isoelectronic sequence. Data points are shown only where theoretical and experimental values exist for the same atomic number. (For references of experiments, see the caption of Fig. 3.) Calculations are from Kim *et al.* (Ref. [14]), Chen *et al.* (Ref. [12]), and Blundell (Ref. [13]).

the Blundell plot the single uranium measurement is represented by three data points. This is because Blundell did not give a value for Z=92 in Ref. [13], and we have used an interpolated QED value by Blundell [32] together with three different RMBPT values. The RMBPT value corresponding to the upper data point was also calculated by Blundell [33] and the lower two are from a calculation by Johnson et al. [34], where two different model potentials as a starting point for their self-consistent calculation were used. The difference between these two values is quoted to give a measure of the convergence of the calculation, since the starting value should not make a difference to the final outcome. In addition to the published calculated values [12-14], interpolations by Blundell for Z=28 and 47 and the QED value for Z=92 [32] and one interpolation by Cheng for Z=47 [30] were used. Nuclear polarization contributions are again omitted from the calculated values, since they are now known to be less than 0.1% of the QED contribution for even the highest Z [31].

One result of Fig. 9 is that for all Z the transition energy values of Kim *et al.* are about 0.6% in QED units smaller than experimental values. Due to the larger error bars and the small number of precise data, no distinction between the other two calculations can be made at intermediate Z. At high Z, the EBIT data [4–6] could indicate a difference between E^{expt} and E^{theor} calculated by Chen *et al.* of about 0.8% in "QED units." The EBIT results for Z=90 and 92 are within the error bars in agreement with Blundell's calculation.

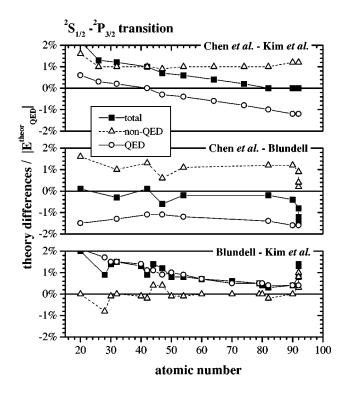


FIG. 10. Normalized differences of the theoretical predictions for the QED contribution, the non-QED part, and the total energy for the $2s^{2}S_{1/2}$ - $2p^{2}P_{3/2}$ transition along the lithiumlike sequence. The calculations were performed by Chen *et al.* (Ref. [12]), Blundell (Ref. [13]), and Kim *et al.* (Ref. [14]).

In Fig. 10 the differences between the three theoretical predictions of QED, non-QED, and the total transition energy are presented for the 1/2-3/2 transition. These differences are normalized to the absolute value of the QED contribution. For the definition of an "experimental QED energy" it is most important to have a reliable non-QED value, but for the 1/2-3/2 transition energy calculations the agreement among calculations is worse than in the 1/2-1/2case. Even though in the non-QED terms excellent agreement between Blundell and Kim et al. is found, the non-OED value of Chen et al. deviates from both of the others for all Z by constantly 1.0% QED units. The situation is especially difficult to judge, since the Chen et al. non-QED calculation is quoted to be more reliable, due to the inclusion of higher-order Breit terms not considered by the other two authors. Also a small dip at the interpolated non-QED value for Z=47 from Blundell [32] is present, suggesting that this interpolated value may be less reliable. A comparison of the total transition energies calculated by Blundell and Chen et al. shows good agreement, except for Z=92. On the other hand, differences of 1% in "QED units" exist for both the non-QED and the QED terms, which nearly cancel each other in the total transition energy. A discussion of the reason for this cancellation is not within the scope of this paper. We conclude that the calculated 1/2-3/2 non-QED terms are not sufficiently accurate for an extraction of QED information out of the measured total transition energies. The need for more precise medium- and high-Z experimental data and calculations is evident.

V. SUMMARY AND CONCLUSIONS

We have performed precise beam foil spectroscopy experiments measuring 2s-2p transition energies in lithiumlike Ag⁴⁴⁺ ions. An improvement in experimental uncertainty by more than one order of magnitude has been achieved over previous beam foil measurements for intermediate Z lithiumlike ions. The experiment is sensitive to two-photon QED processes. For the $2s^2S_{1/2}-2p^2P_{1/2}$ transition, the non-QED terms are well established [12-14]. Thus we can attribute the differences between experiment and calculations to the uncertainty of the calculated OED contributions. Even though in the medium and high Z range few precise experiments exist, we find that the QED terms calculated by Chen et al. show the best agreement with our experiment and other precise data [7,8]. The QED terms evaluated by Blundell include two-photon exchange screening corrections, but do not contain all second-order terms. Therefore deviations from experiment, which show up in the intermediate Z range, are not unexpected.

For the $2s {}^{2}S_{1/2} - 2p {}^{2}P_{3/2}$ transition, disagreements among the non-QED values from different calculations do not allow an unambiguous derivation of an experimental QED contribution. Furthermore, the QED sensitivity of measurements of this transition energy is usually smaller due to the smaller

relative QED contribution. This demands higher-precision experiments in order to achieve the QED sensitivity of the 1/2-1/2 transition energy measurements. The experimental uncertainties are at present of the order as the residual differences between the most advanced calculations.

We conclude that our measurement for the lithiumlike $2s^2S_{1/2} - 2p^2P_{1/2}$ transition energy in Ag⁴⁴⁺, the Doppler tuned energy measurement for the same transition in U⁸⁹⁺, and the EBIT measurement for the 1/2-3/2 transition in Bi⁸⁰⁺ are at present the only measurements which are sensitive to higher-order QED contributions.

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- [1] R. J. Knize, Phys. Rev. A 43, 1637 (1991).
- [2] J. Sugar, V. Kaufman, and W. L. Rowan, J. Opt. Soc. Am. B 9, 344 (1992); 10, 13 (1993).
- [3] J. Reader, J. Sugar, N. Acquista, and R. Bahr, J. Opt. Soc. Am. B 11, 1930 (1994).
- [4] P. Beiersdorfer, Nucl. Instrum. Methods Phys. Res. B 99, 114 (1995).
- [5] P. Beiersdorfer, A. L. Osterheld, S. R. Eliott, M. H. Chen, D. Knapp, and K. Reed, Phys. Rev. A 52, 2693 (1995).
- [6] P. Beiersdorfer, A. L. Osterheld, J. H. Scofield, J. R. Crespo López-Urrutia, and K. Widmann, Phys. Rev. Lett. 80, 3022 (1998).
- [7] J. Schweppe, A. Belkacem, L. Blumenfeld, N. Claytor, B. Feinberg, H. Gould, V. E. Kostroun, L. Levy, S. Misawa, J. R. Mowat, and M. H. Prior, Phys. Rev. Lett. 66, 1434 (1991).
- [8] E. Hinnov and B. Denne, Phys. Rev. A 40, 4357 (1989).
- [9] S. Martin, J. P. Buchet, M. C. Buchet-Poulizac, A. Denis, J. Désesquelles, M. Druetta, J. P. Grandin, D. Hennecart, X. Husson, and D. Lecler, Europhys. Lett. 10, 645 (1989).
- [10] R. Büttner, B. Kraus, K.-H. Schartner, F. Folkmann, P. H. Mokler, and G. Möller, Z. Phys. D 22, 693 (1992).
- [11] U. Staude, Ph. Bosselmann, R. Büttner, D. Horn, K. -H. Schartner, F. Folkmann, A. E. Livingston, Th. Ludziejewski, and P. H. Mokler, Phys. Rev. A 58, 3516 (1998).
- [12] M. H. Chen, K. T. Cheng, W. R. Johnson, and J. Sapirstein, Phys. Rev. A 52, 266 (1995).
- [13] S. A. Blundell, Phys. Rev. A 47, 1790 (1993).
- [14] Y.-K. Kim, D. H. Baik, P. Indelicato, and J. P. Desclaux, Phys. Rev. A 44, 148 (1991).
- [15] A. E. Livingston, R. Büttner, A. S. Zacarias, B. Kraus, K.-H. Schartner, F. Folkmann, and P. H. Mokler, J. Opt. Soc. Am. B 14, 522 (1997).

- [16] W. Persson, C.-G. Wahlström, L. Jönsson, and H. O. Di Rocco, Phys. Rev. A 43, 4791 (1991).
- [17] R. L. Kelly, J. Phys. Chem. Ref. Data 16, 1 (1987).
- [18] J. D. Garcia and J. E. Mack, J. Opt. Soc. Am. 55, 654 (1965).
- [19] V. Kaufman and W. C. Martin, J. Phys. Chem. Ref. Data 20, 775 (1991).
- [20] D. Horn, diploma thesis, University of Giessen (1997).
- [21] K. T. Cheng, Y.-K. Kim, and J. P. Desclaux, At. Data Nucl. Data Tables 24, 111 (1979).
- [22] H. G. Berry, R. W. Dunford, and A. E. Livingston, Phys. Rev. A 47, 698 (1993).
- [23] G. W. F. Drake, Can. J. Phys. 66, 586 (1988).
- [24] R. Marrus, A. Simionovici, P. Indelicato, D. D. Dietrich, P. Charles, J.-P. Briand, K. Finlayson, F. Bosch, D. Liesen, and F. Parente, Phys. Rev. Lett. 63, 502 (1989).
- [25] K. W. Kukla, A. E. Livingston, J. Suleiman, H. G. Berry, R. W. Dunford, D. S. Gemmell, E. P. Kanter, S. Cheng, and L. J. Curtis, Phys. Rev. A 51, 1905 (1995).
- [26] D. R. Plante, W. R. Johnson, and J. Sapirstein, Phys. Rev. A 49, 3519 (1994).
- [27] H. Persson, I. Lindgren, L. N. Labzowsky, G. Plunien, T. Beier, and G. Soff, Phys. Rev. A 54, 2805 (1996).
- [28] P. J. Mohr, Ann. Phys. (N.Y.) 88, 26 (1974); 88, 52 (1974);
 Phys. Rev. A 26, 2338 (1982).
- [29] B. Edlén, Phys. Scr. 19, 255 (1979).
- [30] K. T. Cheng (private communication).
- [31] G. Plunien and G. Soff, Phys. Rev. A 53, 4614 (1996).
- [32] S. A. Blundell (private communication).
- [33] S. A. Blundell, W. R. Johnson, and J. Sapirstein, Phys. Rev. A 41, 1698 (1990).
- [34] W. R. Johnson, J. Sapirstein, and K. T. Cheng, Phys. Rev. A 51, 297 (1995).

- [35] J. H. Davé, U. Feldman, J. F. Seely, A. Wouters, S. Suckewer, E. Hinnov, and J. L. Schwob, J. Opt. Soc. Am. B 4, 635 (1987).
- [36] A. S. Zacarias, A. E. Livingston, Y. N. Lu, R. F. Ward, H. G. Berry, and R. W. Dunford, Nucl. Instrum. Methods Phys. Res. B 31, 41 (1988).
- [37] R. Büttner, doctoral thesis, University of Giessen (1997).
- [38] W. E. Behring, J. F. Seely, C. M. Brown, U. Feldman, and J. P. Knauer, J. Opt. Soc. Am. B 6, 531 (1989).
- [39] D. D. Dietrich, J. A. Leavitt, H. Gould, and R. Marrus, Phys. Rev. A 22, 1109 (1980).
- [40] S. Martin, A. Denis, M. C. Buchet-Poulizac, J. P. Buchet, and J. Désesquelles, Phys. Rev. A 42, 6570 (1990).
- [41] R. Büttner, B. Kraus, M. Nicolai, K.-H. Schartner, F. Folkmann, P. H. Mokler, and G. Möller, in the *VIth International Conference on the Physics of Highly Charged Ions*, edited by P. Richard, M. Stoekli, C. L. Cocke, and C. D. Lin, AIP Conf. Proc. No. 274 (AIP, New York, 1993), p. 423.