potential maximum does not represent a barrier to zero-energy collisions with zero angular momentum.

The cross section for the formation of positronium and protium pairs, calculated using the adiabatic model together with classical trajectories, is shown in Fig. 2. The cross section decreases monotonically from  $31a_0^2$  at 0.01 eV to  $4a_0^2$  at 20 eV. These results should be accurate over the higher energies in this range, but quantum effects on the nuclear motion may influence the results at low energies.

The maximum in the H- $\overline{H}$  potential is probably a feature common to all interaction potentials between neutral atoms and antiatoms. This suggests the possibility that for some pairs the potential at the maximum might be greater than the dissociation energy, thus providing a potential barrier which could keep matter and antimatter apart. This could perhaps give some support to the recent speculation<sup>6</sup> that ball lightning is caused by the congregation of large amounts of antimatter in the atmosphere. Preliminary studies on He- $\overline{H}$ , with 7-term wave functions, reveal a small barrier which will probably disappear in more accurate calculations. Our results for systems in which one or other atom is charged show no potential barriers.

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## Metastable Autoionizing States of Highly Excited Heavy Ions\*

I. A. Sellin, D. J. Pegg, and P. M. Griffin

University of Tennessee, Knoxville, Tennessee 37916, and Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

and

## Winthrop W. Smith

University of Connecticut, Storrs, Connecticut 06268 (Received 20 March 1972)

We report the respective electronic energies  $[-5.658 \pm 0.024 \text{ and } -6.369 \pm 0.033 \text{ keV}]$ and electron emission lifetimes  $[0.91 \pm 0.04 \text{ and } 0.66 \pm 0.04 \text{ nsec}]$  of the longest-lived autoionizing quartet states of lithiumlike  $Cl^{14+}$  and  $Ar^{15+}$  ions, firmly establishing the energy and lifetime scaling with Z. We discuss observable relativistic corrections to nonrelativistic variational energy calculations. Production of metastable states belonging to lower charge states is discussed, and it is concluded that many excited heavy ions should exhibit them.

Sufficient numbers of metastable Cl<sup>14+</sup> and Ar<sup>15+</sup> ions have been created by passage of chlorine and argon beams through  $15-\mu g/cm^2$  carbon foils to render possible the measurement of the total electronic energies and autoionization lifetimes of the lowest, longest-lived, nonradiative quartet states [ $(1s2s2p)^4P_{5/2}^{\circ}$ ] of these three-electron ions. 41-MeV chlorine beams from the Oak

Ridge tandem accelerator and 87- and 60-MeV argon beams from the Oak Ridge isochronous cyclotron were used for these experiments. In each case a few tenths of 1% of the emergent three-electron beam fraction has been found to exist in the lowest quartet state having highest angular momentum  $(J = \frac{5}{2})$ ; it is then probable that  $\gtrsim 1\%$  of the three-electron beam fraction is metastable to some degree, when other J states are considered, and the beam energy is adjusted for optimum yield.<sup>1</sup> Moreover, we have observed production of metastable autoionizing states belonging to other chlorine charge states, which also presumably have high spin. Hence the experiments reported here provide new evidence that production of metastable autoionizing states is a common feature of the penetration of energetic heavy ions in matter.

An electron spectrometer is used to study the decay in flight of energy-selected electrons spontaneously emitted by the excited beam, as a function of the separation of the target from the spectrometer viewing region. The apparatus and techniques used by us are fully set forth in Ref. 1, together with an assessment of experimental errors. The most important of these stem from uncertainties in the beam velocity (because of the imperfectly known energy loss in the target) and angular spread in the electron emission direction (which enters into the kinematic transformation connecting electron velocities in the laboratory and ionic rest frames). The electron spectrometer resolution (including kinematic spread) in the present work was 0.6% full width at half-maximum.

The segment of the electron emission spectrum pertaining to the lowest quartet state of the  $Cl^{14+}$ ion is shown in Fig. 1. A single peak is seen, in contrast to our earlier work<sup>1</sup> with oxygen and fluorine beams. This observation is consistent with the expectation that the adjacent features re-

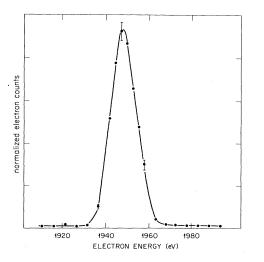


FIG. 1. Segment of the electron spectrum from 41-MeV chlorine ions undergoing decay in flight. The peak arises from the lowest three-electron quartet state, and is plotted in the ionic rest frame.

ported in Ref. 1 are too short-lived for Z = 17 to make their appearance at the prevailing targetspectrometer separation of 3 cm. The single feature is then associated with the decay of the  $(1s2s2p)^4P_{5/2}^{\circ}$  state of the  $Cl^{14+}$  ion, on the basis of the known metastability<sup>2</sup> of analogous states in isoelectronic systems of low Z, and because the metastable state energies deduced correspond closely to well-founded extrapolations of the calculations of Holøien and Geltman<sup>3</sup> for Z = 4 to 10. The same situation prevails for a corresponding peak we have observed in the  $Ar^{15+}$  electron spectrum.

There are, however, small differences between the anticipated electron energies and those experimentally observed, which are presumably due to purely relativistic effects. The observed electron energies correspond to the difference in energy between the initial  ${}^{4}P^{\circ}$  state, and the final state, which consists of the  ${}^{1}S_{0}$  heliumlike ion in its ground state plus a continuum electron having suitable angular momentum and parity  $({}^{2}F_{5/2}^{\circ})$ . Hence the electron emission energy minus the sum of the well-established one- and two-electron ion ionization potentials<sup>4</sup> yields the  ${}^{4}P^{\circ}$  state energy directly. The experimental state energies are thus  $-5.658 \pm 0.024$  keV and  $-6.369 \pm 0.033$  keV for Cl<sup>14</sup> and Ar<sup>15+</sup>, respectively. These experimentally derived energies can be compared with the extrapolated nonrelativistic variational calculations by Holøein and Geltman,<sup>3</sup> suitably corrected for relativistic effects. The extrapolation was carried out by fitting by least squares a power series in decreasing powers of Z (beginning with  $Z^{2}$ ) to the energies in Ref. 3, a nonrelativistically rigorous procedure.<sup>5</sup> For Z = 17, 18 the convergence is rapid, and the extrapolated  ${}^{4}P^{\circ}$  state energies are - 5.659 and - 6.362 keV, respectively. The leading relativistic correction to these energies stems from the relativistic mass increase of the single K-shell electron,<sup>6</sup> which in the absence of outer electron screening effects amounts to  $-\frac{5}{4}\alpha^2 Z^4$  Ry, or -75 eV for Cl<sup>14+</sup> and -95 eV for  $\text{Ar}^{15+}$ . Inclusion of this relativistic correction alone then removes the overlap of experimental and theoretical energies. It thus seems reasonable to suppose that a portion of the increased binding is compensated by other relativistic corrections, of which the largest are likely to be the Dirac electric-moment, spin-orbit, and spin-spin corrections. To our knowledge these have not been worked out in detail; when detailed calculations are available, it would be worthwhile and possible to refine the experimental values for comparison.

Recent results on lifetimes of the  $(1s2s2p)^4P_J^{\circ}$ states for low-Z systems have been reviewed by Feldman, Levitt, and Novick,<sup>7</sup> Estberg and La-Bahn.<sup>8</sup> and in Ref. 1. We briefly recall that Coulomb autoionization is forbidden for these states (as is electric dipole radiation) because of the required spin change. For  $J = \frac{5}{2}$ , the spin-orbit matrix elements also vanish, so that the lifetime of the  $J = \frac{5}{2}$  state directly measures the spin-spin-interaction matrix element. The addition of values for the  ${}^{4}P_{5/2}^{\circ}$  lifetime for Cl<sup>14+</sup> and Ar<sup>15+</sup> in this work considerably extends the range of measured values along the isoelectronic sequence from Z= 9 to Z = 17 and 18, for which we measure 0.91  $\pm 0.04$  nsec and  $0.66 \pm 0.04$  nsec, respectively. To our knowledge there are no theoretical values with which to compare the latest results, nor can we comment on possible explicitly relativistic effects in the lifetime calculations. Levitt, Novick, and Feldman<sup>9</sup> have suggested the approximate semiempirical lifetime scaling law  $(Z - 1.75)^{-3}$ ; however, with the addition of the values for Z=17,18 it is clear that this choice is inadequate. In fact, the form  $a(Z-b)^{-c}$  provides only a mediocre fit to the  ${}^{4}\!P_{5/2}$  lifetime data over the range Z = 2-18. In any case, the "best" value for c, which we determined by a least-squares fit to the available data for Z = 2, 3, 8, 9, 17, and 18, is about 3.5 rather than 3. The fit to the quantity bis, of course, insensitive to the high-Z data, so no result is quoted. The lifetime measurements for Z = 4-8 of Dmitriev, Nikolaev, and Teplova<sup>10</sup> are not fitted, because of discrepancies with our earlier oxygen measurement.

The measured lifetime values of  $0.91 \pm 0.04$  and  $0.66 \pm 0.04$  nsec were obtained by the methods discussed in Ref. 1, in which experimental details may be found. The standard method of measuring the electron counting rate (per unit beam current) versus target position was used to track the  ${}^{4}P_{5/2}^{\circ}$  state decay in flight, over about three decay lengths, for both Cl<sup>14+</sup> and Ar<sup>15+</sup> ions. Figure 2 shows decay-in-flight data obtained with a 60-MeV argon beam. This curve has been corrected for a small background, amounting to  $\lesssim 3\%$ of the total intensity at the upstream data points. Most of this background was due to beam-independent detector noise, but for the argon data (though not for chlorine) there was a measurable scattered electron background in the wings of the spectral peak. Uncertainties in this beam-dependent background are responsible for the quoted Ar<sup>15+</sup> error being greater than that for Cl<sup>14+</sup>. For

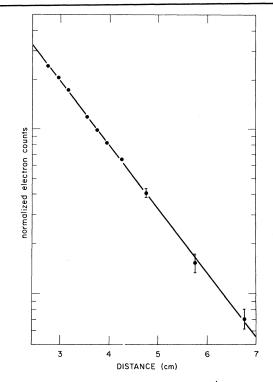


FIG. 2. Decay in flight of the  $(1s2s2p)^4P_{5/2}^{\circ}$  state of Ar<sup>15+</sup> ions, obtained at a cyclotron beam energy of 60 MeV.

points showing no error flags, the statistical counting errors were smaller than the symbol diameter. For both the Cl<sup>14+</sup> and Ar<sup>15+</sup> data, least-squares-fitted lifetimes were examined as a function of the number of data points retained. Removing up to four points from the upstream and later from the downstream portions of each decay curve caused the fitted lifetimes to oscillate about the quoted values, with a range error substantially smaller than the quoted error limits. Both the statistical errors given by the leastsquares program and the range error we have described are well within the quoted error. Reproducibility from run to run was also within this error. Uncertainties in the possible foil-aging effects we have encountered previously<sup>11</sup> are then partly responsible for the error limits we have chosen.

In Ref. 1, evidence for the production of metastable autoionizing states of oxygen and fluorine ions having four or more electrons was presented (for berylliumlike ions, quintet spin states would be the most metastable). We have now observed such states in chlorine ions having substantially more than four electrons; but the energies, assignments, and lifetimes of these states are still under experimental investigation and will be reported elsewhere.

What we conjecture is that such states are commonly formed during the penetration of energetic heavy ions in matter, and that in situations where mean autoionization lifetimes are of interest, it is by no means safe to assume that essentially all energetically permitted autoionization transitions occur in a time  $\le 10^{-14} \text{ sec.}^{12}$ The whole subject of the role of metastable autoionizing states in the penetration of energetic heavy ions in matter was largely ignored by Bohr and Lindhard<sup>13</sup> in their 1954 monograph on electron capture and loss by heavy ions penetrating through matter, although this omission was duly noted by them. On the balance we have no reason to suspect any gross failure in the Bohr-Lindhard (BL) theory, since the metastable production we have found thus far is far from dominant. On the other hand, our data tend to support the arguments of Bohr and Lindhard that the average level of excitation is high; for example, with the  $Ar^{15+}$  beams used in this work the  ${}^{4}P^{\circ}$  state lies about 2.2 keV above the ground state of  $Ar^{16+}$ , to which it decays. This is only one of many possible highly excited states. The modifications proposed by Betz to the BL theory require a much lower average excitation than in the BL theory. Hence the observation of a number of metastable states involving substantial emission of electrons in the range of 100 eV to several keV tends to favor the Bohr-Lindhard model.

It is plausible that formation of a substantial number of long-lived and intermediate-lived autoionizing states should occur during the penetration of energetic heavy ions through matter. Consider a subshell with a single hole in it deep within the ion. Because of the Pauli principle, the filling of this hole requires that the spin of the electron in question have the correct orientation. If the total electronic spin of the excited state prior to autoionization (assuming LS coupling) is inconsistent with the spin of the only residual ion state permitted, then the excited state is automatically stable against Coulomb autoionization, but can in general autoionize through the much weaker spin-orbit and spin-spin interactions. Hence the question of the formation of metastable autoionizing states has to do with the statistical distribution of the electronic spin states of excited ions formed by electron capture. Many excited configurations belonging to a wide variety of charge states should exhibit this behavior.

Finally, we note that the copious formation of a number of metastable excited states of highly charged heavy ions indicates a bright future for experiments devoted to the atomic structure and lifetimes of such systems, which are rare on earth but have high astrophysical significance.

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