A Stored-Ion Target for X-ray Spectroscopy of Multicharged Ions

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Abstract

With the evolution of the new third generation synchrotron radiation sources providing intense beams of hard x-rays, it is natural to consider exploiting these to investigate one of the central problems in atomic and molecular physics: the 3-body Coulomb problem. The atomic physics community could advance this field considerably by developing general techniques to investigate the x-ray spectroscopy of heliumlike ions. To do so, however, requires the development of a target of such ions with sufficient density to permit photoexcitation studies in the hard x-ray regime. A possible scheme to achieve this is described. Such a target system would permit x-ray investigations with exotic species such as highly-charged atomic ions, size-selected cluster ions, and atomic and molecular negative ions which have hitherto been impractical to study with conventional techniques.

One of the current frontiers of atomic physics is in understanding atomic structure in the regime where relativistic effects and electron-electron correlations are simultaneously important.[1] A good testing ground for this regime is the study of heliumlike ions at intermediate Z. In particular, it

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has long been recognized that doubly-excited states of heliumlike ions provide a unique handle on electron-electron correlations just by the very fact that they can be reached by photoexcitation from the ground state despite the single-particle nature of the photon-electron dipole operator.[2] Furthermore, because such systems deviate so strongly from the independent particle approximation, they are governed by very different quantum numbers and selection rules from single-particle states[3] leading to spectral patterns more closely-related to molecular spectra than atoms.[4]

For helium (and heliumlike ions), the energy of any state in which both electrons are excited is higher than the ionization threshold and thus such states appear as autoionizing continuum resonances. The autoionization continuum of He has been studied in great detail for over 30 years. In fact, the pioneering work of Madden and Codling[2] on the doubly-excited autoionizing states of He is generally considered to mark the start of the Synchrotron Radiation era in atomic physics.[5] Despite this long history, and the great interest in the 3-body Coulomb problem,[6] there is little experimental information available on such states in systems heavier than helium. Beam-foil experiments[7] have observed some doubly-excited states in heliumlike ions up to oxygen, though the only high-resolution data that exists on heavier systems comes from dielectronic recombination measurements in heavy-ion storage rings.[8] While there have been several recent theoretical attempts to predict the Z-scaling for double photoionization[9], there are no data available with which to test such theories. The fact remains that despite more than 30 years of effort in this field, long "families" of doubly-excited states have only been observed in H\(^-\) and even then only for limited excitation regions.[10]

The importance of extending these investigations to heavier members of the helium isoelectronic sequence is simple to understand by looking at the form of the perturbative term employed in the Hylleraas-Scherr-Knight variational procedure[11]. In this commonly-used perturbative method[12], after scaling energy and length by \(Z^2\) and \(Z^{-1}\) respectively, the nonrelativistic 2-electron Hamiltonian is written in the form:

\[
H = H_0 + Z^{-1}V
\]

where \(H_0\) represents the summed hydrogenic Hamiltonians of the two electrons and \(V = 1/r_{12}\) is the perturbative electron-electron term. In the limit \(Z \rightarrow \infty\), the correlation term vanishes and the independent particle approx-
imation would become exact. However, with increasing \(Z\), relativistic interactions become increasingly important with terms growing as \(Z^2\) and \(Z^4\).[1] Thus, the ability to experimentally follow trends with \(Z\) in the heliumlike sequence would provide theorists an important new tool.

Third-generation synchrotron radiation sources such as the Advanced Photon Source present us with a remarkable opportunity to address this problem because of the high brilliance which will be available. Though the investigation of heliumlike ions is the principal motivation behind the idea of developing a stored ion target for use at the APS, such a facility would be far more general. Given suitable ion sources, such a stored ion target would represent a major breakthrough for synchrotron-related research with other exotic targets such as negative ions and cluster ions as well. Thus, the success of this project would considerably enhance the capabilities of researchers in several different fields.

Perhaps the biggest challenge in carrying out these experiments is making sufficient numbers of heliumlike heavy ions and then constraining them in a high-density region to interact with the photon beam in a cost-effective manner. Several groups around the world are either developing, or have already constructed, facilities for photon-ion merged beam experiments with synchrotron radiation (see e.g. [13] and references therein). These are all variants of the apparatus for photoion spectrometry on singly-charged ions pioneered by West and his collaborators at Daresbury.[14] At least two of these groups plan to upgrade these single-pass systems to study multiply-charged target ions by the addition of Electron Cyclotron Resonance (the ICARIES project at Super ACO[13]) and Electron Beam Ionization Source (MERGING at Spring-8 [15]) ion sources.

None of the single-pass apparatuses however will produce sufficient densities of heavy heliumlike ions for spectroscopic studies. We propose instead to accomplish this by means of ion trapping techniques. Such methods can provide long interaction times with the radiation over extended interaction lengths thus compensating for the lower densities in comparison to neutral targets. Previously, synchrotron studies with ion traps have employed stationary ions in small localized structures such as Penning traps.[16]. In such experiments, charge state distributions have been limited by charge-changing with the residual gas in the trap. This problem can be suppressed considerably by forming the ions external to the trap and by introducing a relative velocity between the ions and the stationary neutral background gas in a storage ring geometry.
For example, one promising method currently being considered utilizes a “racetrack” storage ring[17, 18] (see Fig. 1). In this geometry, ions circulate in a closed loop rf-quadrupole (RFQ) filter[19] designed with long (~ 1 meter) backstretches for interaction and probing regions. This could serve both to form the heliumlike ions by photon (and possibly also electron) impact of few-times ionized ions injected from inexpensive rf- or Penning (PIG) sources.

The basic operating characteristics of a quadrupole mass filter derive from the motion of a particle in a rotating 2-dimensional quadrupole field.[20] In an electrostatic quadrupole, a charged particle is bound in one direction (x in Fig. 2) though unbound in the transverse direction (z). By adding a time-dependent rf component, this saddle potential rotates to exchange these directions and consequently leads to a time-averaged parabolic pseudopotential forming a 2-dimensional harmonic trap which can be used to bind particles to the rotation axis. The equations of motion in the x – z plane then take the form of Mathieu equations[21] which can be written:

\[
\frac{d^2x}{d\tau^2} + (a + 2q \cos 2\tau)x = 0,
\]

\[
\frac{d^2z}{d\tau^2} - (a + 2q \cos 2\tau)z = 0
\]

in terms of the dimensionless parameters

\[
a = \frac{4QU}{Mr_0^2\Omega^2}, \quad q = \frac{2QV}{Mr_0^2\Omega^2}, \quad \text{and} \quad \tau = \frac{\Omega t}{2}
\]
Figure 2: Saddle potential formed by electrostatic quadrupole field.

where $U$ and $V$ are the dc and rf field strengths respectively, $r_0$ the radius of the quadrupole field, and $\Omega$ the rf drive frequency. These equations have both stable and unstable solutions. The question of stability depends only upon the parameters $a$ and $q$, which in turn depend upon the mass to charge ratio of the ion ($M/Q$) but not on the initial conditions such as velocity. Consequently, the $a - q$ space consists of regions of simultaneous stability separated by regions of $x-$ and/or $z-$instability (see Fig. 3). For fixed values of the field parameters ($U$, $V$, and $\Omega$), all particles with the same $M/Q$ will be trapped by a unique combination of $a$ and $q$ known as the operating point. Because the ratio $a/q (= 2U/V)$ is independent of $M$ and $Q$, all masses and charges will lie along an operating line with fixed $a/q$. For fixed mass, the charge states are dispersed along this line (Fig. 3) with neutrals at the origin up to a maximum charge state determined by the parameters. Because of the triangle-like shape of this region of stability, when operated with a low value of $a/q$ (small dc component), all charge states (up to some maximum) can be trapped simultaneously. However, as this ratio increases toward the limiting value 0.336, the trap becomes increasingly selective and can trap a single charge state. Thus, by slowly ramping the dc voltage $U$ from zero, the ring can be changed in character from a broad acceptance with all charge states present to a highly selective filter with a unique charge state confined.
Figure 3: Lowest region of stability for the RFQ trap as a function of $a$ (the dc field strength) and $q$ (rf field strength). Three typical operating lines are shown with respective charge states of Kr up to 36+ indicated. In the broadband filter mode ($a/q=.02$), all charge states are trapped simultaneously while for higher operating lines, trapping becomes increasingly selective.

In a linear RFQ mass filter, ion motion along the longitudinal axis ($y$) is free. In the storage ring geometry however, a centripetal force is supplied in the bend regions (radius $R$) by the radial component of the pseudopotential and consequently, the field and geometric parameters determine a maximum kinetic energy $E_{\text{max}}$ that can be confined by the storage ring given by:

$$E_{\text{max}} = \frac{QV R q}{16 r_0}.$$

Finally, we consider the maximum ion density which can be confined. This can be estimated by comparing the space charge potential of a closed cylinder of radius $r_0$ to the value of the time-averaged pseudopotential on
that cylinder. [19] This yields the limiting density

\[
\rho_{\text{max}} = \frac{V^2}{4\pi M r_0^4 \Omega^2}.
\]

This is of course a theoretical limit, and in practice most experimenters have usually operated at lower densities.[18] Such densities could however be approached with improved mechanical structures.[22] In fact, Walther and collaborators recently employed a circular RFQ ring to trap sufficient ion densities to observe ordered ion structures.[23]

As an example, consider the case of 20-keV Kr^{34+} in Fig. 3. Such ions could be accumulated up to a theoretical space-charge limit of \(10^{10}\) total particles (at a density of \(10^8/\text{cm}^3\)). With an orbital period of \(4 \times 10^4/\text{sec}\), this produces an effective current of 70 particle-microamperes. Assuming a photon brilliance[24] of \(10^{17}\) photons/\text{sec}/0.1\%BW/mrad²/mm², the expected luminosity for photon-ion interactions would be \(\mathcal{L} = 2 \times 10^{26}/\text{sec} \cdot \text{cm}^2\).

Based on practical experience however, we would expect to achieve only 1-2 orders of magnitude less density than this theoretical limit because of field imperfections.[25] Resonances as weak as \(5 \times 10^{-22}\) cm² could be observable with a production rate in excess of \(10^3\) per second and thus there provide sufficient signal for a wide variety of experiments involving realistic atomic cross sections which are generally several orders of magnitude larger.[9] Furthermore, with the possibility of employing non-destructive analog resonant detection techniques that are practical in such traps,[18] near-unit detection efficiency should be achievable for experiments involving ion detection.

By developing such a system to study the interaction of x-rays with highly-charged ions, we will have a unique opportunity not only to observe doubly-excited states in intermediate-Z ions but, for the first time, to follow the trends of the doubly-excited states in iso-electronic 2-electron systems. This would provide the first truly comprehensive test of the various theoretical descriptions of these basic 3-body Coulomb systems that are central to many fields of physics and chemistry.[10] Furthermore, such studies could naturally be extended to the so-called “triply-excited” states in 3-electron ions which have recently been reported in lithium.[26] These lithium-like systems provide the opportunity to excite both doubly-excited bound and autoionizing states and follow the ionization threshold with increasing atomic number.

Such a novel apparatus will be extremely promising for a variety of other interesting experiments with ionic targets. For example, complementary to our proposed studies of helium-like positive ions, this facility would also be
adapted to study negative alkali ions, which are analogous to the 2-electron ions in terms of electron correlation phenomena. With the addition of a Cs sputter source for negative ions, similar techniques could be used to investigate such species.

Other proposed uses for such a facility include high resolution spectroscopy of hydrogenlike species by photoexcitation. In contrast to existing emission spectroscopic investigations of such species which are plagued by weak signals, we could take advantage of the monochromaticity of the incident beam and exploit efficient non-dispersive detection techniques. In addition, the accumulation and storage of size-selected cluster ions would, permit studies of photofragmentation with tunable size-selected beams.

We believe that such a facility, because of its unique characteristics, will open up a wealth of new areas of research which have hitherto been either difficult or completely inaccessible.

References


