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Positive Ion Properties from Spectroscopy of High-L Rydberg levels with the RESIS Method¹

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All atoms and ions have many singly-excited levels with large values of angular momentum ($L > 5$). The existence of these nearly hydrogenic levels plays an important role in dynamic processes, but the details of their binding energies are often ignored since they correspond to very small quantum defects ($\delta < 0.001$) and are very difficult to observe with standard spectroscopic methods. One very general method that has been developed specifically to explore the spectroscopy of these high-L levels is the RESIS, or Resonant Excitation Stark Ionization Spectroscopy method. With this technique, high-L Rydberg levels formed by charge capture in a fast atom or ion beam are resonantly excited upwards by a laser, and the upper level so populated is then Stark Ionized and the resulting ion collected with high efficiency. Because the laser transition is upwards, selection rules do not limit the angular momentum of detected levels, and many different high-L levels can be detected, resolved by the small differences between their excitation energies and the hydrogenic transition energy. Once selectively detected in this way, RF/Optical double resonance methods can measure the binding energy differences between adjacent levels with high precision. The binding energies of these high-L levels are a sensitive indicator of many properties of the positive ion binding them, such as polarizabilities and permanent electric moments. Since these properties are otherwise difficult to measure and can be difficult to calculate with confidence, the information derived from RESIS spectroscopy can provide tests of advanced theoretical methods and input to applications involving long-range interactions of atoms or ions. Two recent studies illustrate the method. One determined the dipole and quadrupole polarizabilities of the Rn-like Th⁴⁺ ion by measuring the binding energy differences between $n=37$ Rydberg levels of Th³⁺ with $L=8-15$ [1]. Another determined the quadrupole and hexadecapole moments and dipole and quadrupole polarizabilities of the Fr-like Th³⁺ ion by mapping out the complex pattern of binding energies of $n=28$ Rydberg levels of Th²⁺ with $L=9-12$ [2].

[1] Julie A. Keele, Chris. S. Smith, and S.R. Lundein, Phys. Rev. A **85**, 064502 (2012)

[2] Julie A. Keele, Chris S. Smith, and S.R. Lundein, Phys. Rev. A **88**, 022502 (2013)

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