Cold highly charged ions for highest precision spectroscopy

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Highly charged ions (HCI) are ubiquitous in the universe. Their emission spectrum encompasses the range from the visible to the x-rays, and their spectral lines are key elements of astrophysical and fusion plasma diagnostics. However, their spectroscopic study at high resolution has been hindered by the lack of an efficient technique to cool them down by more than six orders of magnitude, namely from typical laboratory production temperatures in the MK range down to a regime in which the Doppler broadening is strongly suppressed. For this, as well as for other technical reasons, there is a gap spanning ten orders of magnitude between the photon energy accuracy which is now achievable in frequency metrology by means of laser spectroscopy with neutral atoms and singly charged ions, and the one found in HCI research. By applying sympathetic cooling inside a Coulomb crystal, we recently overcame this limitation [1-3], preparing them for high-precision laser spectroscopy, as shown in Figs. 1 and 2.

A very important application which has been made possible in this way is the investigation of forbidden optical transitions with much enhanced sensitivity to a variation of fundamental constants that has been highlighted in many recent theoretical works [4-9]. The ground state configurations of HCI belonging to many isoelectronic sequences show fine structure and hyperfine splitting giving rise to forbidden optical transitions suitable for laser spectroscopy and optical clocks.

Very interesting examples of forbidden lines are found near charge-state dependent crossings between the 4f and 5s electronic levels [4]. Due to the near degeneracy between levels of opposite and equal parity, a great variety of forbidden electronic transitions appears. Among them, some of them have been theoretically predicted to possess the conceivable largest relative change in optical frequency as a function of the value of the fine-structure constant α found in an electronic transition. We separately study HCI with such transitions in order to identify them [10].

A further application of HCI currently under development is their use as optical frequency standards for metrology. Their great advantage for this purpose is the extremely reduced susceptibility to external perturbations such as black body radiation induced shifts, since the polarizability of the optically active electron is reduced in HCI by many orders of magnitude in comparison with neutral atoms and singly charged ions.

Since HCI have high ionization potentials, they can be exposed to even to x-ray photons without becoming photoionized and changing charge state by very fast Auger processes. This makes them suitable as future frequency standards beyond the vacuum ultraviolet, a regime where the only alternative would be the excitation of a few, still poorly known nuclear transitions. In contrast, HCI have a huge number of both forbidden and allowed transitions up to the keV range which would be appropriate for frequency metrology at such energies. We are currently developing a frequency comb for the vacuum ultraviolet region to test these possibilities.

![Figure 1: Cryogenic ion trap for sympathetic cooling of HCI. A laser-cooled Be\(^{+}\) Coulomb crystal prepared inside the trap cools hot HCI produced in a separate electron beam ion trap. These are first decelerated and perform oscillations passing through the Coulomb crystal, whereby they lose enough kinetic energy to finally become embedded in the crystal.](image)

Figure 2: Image of a Coulomb crystal of Be\(^{+}\) ions with three Ar\(^{15+}\) ions implanted in its left side. The length of the crystal is approximately 0.5 mm. Due to the strong repulsion exerted by the HCI, the space surrounding them is devoid of laser-cooled Be\(^{+}\) ions and appears darker than the other parts of the crystal. Once implanted, the initially much hotter HCI quickly reach thermal equilibrium with the Coulomb crystal at mK temperatures [1,2], and become spatially localized.

References

Precision atomic spectra calculations for highly-charged ions and electron-hole transitions

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Several recent optical atomic clock proposals have necessitated new methods of calculation of atomic spectra and properties in highly-charged ions and systems where holes play an important role [1]. The potential new clocks are predicted to have extraordinarily high accuracy [2] and reduced systematic effects [3]. In cases where the transitions are available due to level crossings, the clocks can have extremely high sensitivity to variation of the fine-structure constant \( \alpha \), potentially improving current limits on time-variation of \( \alpha \) by up to two orders-of-magnitude.

The experimental spectroscopy of one such candidate, the \( \text{Ir}^{17+} \) ion which has two holes in the otherwise closed \( 4f^{14}5s^2 \) valence shells, has shown that current theoretical methods have severe limitations in accurately describing the spectrum [4]. That study included (along with the experimental spectrum) the results of several calculations including different variants of configuration interaction (CI), multiconfigurational Dirac-Fock, and Fock-space coupled cluster. None of the theories tested were able to unambiguously identify the entire observed spectrum. Furthermore many existing methods of calculation – such as the combined configuration interaction and many-body perturbation theory (CI+MBPT), correlation potential methods, and coupled-cluster methods – are designed to work well in one- or two-valence-electron atoms and particularly in near-neutral systems.

We have developed an \textit{ab initio} method of calculating atomic spectra and properties in systems where holes play an important role. Based on the CI+MBPT method [5], we have implemented Wick contractions numerically in AMBiT allowing the inclusion of configurations with arbitrary numbers of valence holes and electrons. The method can treat valence-hole systems like \( \text{Ir}^{17+} \), electron-hole excitations in, e.g. noble gases, and can be used to add important hole configurations to improve the accuracy of transitions of valence electrons. As a first test case, we have performed calculations of spectra and sensitivity to \( \alpha \)-variation for the \( \text{Hg}^{+} \) ion, where the clock transition \( 6s \rightarrow 5d^{-1}6s^2 \) has been compared with an \( \text{Al}^{1+} \) clock to get the best current limit on time-variation of \( \alpha \) [6]. We present results of the full CI+MBPT method with holes, and updated limits on time-variation of \( \alpha \) based on the existing experiment [7].

References


[6] T. Rosenband \textit{et al.}, Frequency Ratio of \( \text{Al}^{+} \) and \( \text{Hg}^{+} \) Single-Ion Optical Clocks: Metrology at the 17th Decimal Place, Science \textbf{319}, 1808 (2008).

X-ray measurements in exotic atoms increase discrepancy in QED tests

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Recent results on hydrogenic [1-3] and helium-like medium-Z systems [4] are complementary to tests on hydrogen [5] and muonic hydrogen [6] and sensitive enough to discover unexpected new physics. In particular, QED theoretical uncertainties are comparable to experimental accuracy. Recent results at Electron Beam Ion Traps have reduced uncertainty to the 10 ppm region, with calibration standards defined to well below this. This yields surprisingly unexpected results and demonstrates that tests of quantum electrodynamics, QED, in complementary regimes, is crucial to our development of understanding just as recently the investigations of hydrogen and muonic hydrogen have been juxtaposed to world-wide interest. The discrepancy revealed by researchers in Australia, NIST and Hungary [7] had 5-6 s.e. significance. Other recent speculation has suggested that this might be primarily in the higher-Z region, or that it is contraindicated for Argon. The evidence for and against these hypotheses are interesting, as is discussion of the magnitude of incomplete correlation terms in the computations.

Here we report a significant discrepancy in helium-like titanium intercombination and forbidden transitions (x, y, z) in trapped Ti20+ ions [8,9]. Our 15 ppm accuracy is able to discriminate between available QED formulations and reveals a pattern of discrepancy of almost 6 standard errors of experimental results from the most recent theoretical predictions with a functional dependence proportional to Zn where n ≃ 4. In both the muonic and highly charged systems, the sign of the discrepancy is the same, with the measured transition energy higher than predicted. This may give insight into effective nuclear radii, the Rydberg, the fine-structure constant or unexpected large and unaccounted-for higher order QED terms. It also invites a critical consideration of correlated and uncorrelated errors in experimental measurement.

References


![Figure 1: Discrepancy of experimental data from latest theory for two-electron systems.](image1)

![Figure 2: Comparison of current status of two key discrepancies of QED-sensitive measurements from accepted energy determinations, divided by the magnitude of the energy measured. The scales in different types of experiments are quite different. The error bar shown on each point is the standard error of the measurement. The label gives the ratio of the discrepancy to the uncertainty.](image2)
Chaotic compound states in electronic, photonic and atomic processes

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We developed a statistical theory for finite Fermi systems (atoms, molecules, nuclei) based on properties of chaotic eigenstates which are formed when several particles are excited [1–4]. Level density of many-body states exponentially increases with the number of excited particles. When residual interaction exceeds the interval between these levels, the eigenstates (compound states) become a chaotic superposition of of thousands, or even millions of Slater determinant basis states. “Exact” calculations of such eigenstates (compound states) are impossible in principle since all minor perturbations (e.g., higher-order correlations or relativistic effects) are enhanced due to exponentially small energy denominators, and strongly affect the eigenstates.

This situation takes place in highly excited nuclei, rare-earth and actinide atoms [3] and open f-shell ions excited by the electron recombination [5]. Chaotic compound resonances have been recently found in ultracold collisions of erbium atoms [6]. In this case physical problems are solved using statistical theory which predicts observables averaged over a small energy interval containing many compound states.

We applied the statistical theory to calculate orbital occupation numbers, matrix elements, enhancement of weak interactions and electromagnetic amplitudes between chaotic compound states. Our predictions of the $10^{-8}$ - $10^{-9}$ enhancement of the parity violation effects in neutron-nucleus reactions near chaotic p-wave compound resonances [1, 7] have been confirmed by numerous experiments (see, e.g. review [2] and references therein). Our calculations of the electromagnetic transition probabilities between chaotic excited states in Ce atom agree with the experimental data.

We derived formulas for the resonant multi-electron recombination via di-electron doorway states leading to the many-electron compound resonances [4, 5] and performed numerical calculations for the electron recombination with gold ($Au^{25+}$) [5] and tungsten ions ($W^{q+}$) [8, 9]. Our calculations [5, 8, 9] agree with the experimental results for $Au^{25+}$ and $W^{20+}$. The experiment showed that the electron recombination of tungsten ion $W^{20+}$ exceeds the direct recombination by three order of magnitude. Recently two new measurements for $W^{18+}$ and $W^{19+}$ have been published [10]. They also have demonstrated low energy enhancements in recombination rate. We have applied our statistical theory to open f-shell target ions $W^{q+} (4d^{10}4f^n)$, where $q = 25, 24, 23, 22, 21, 20, 19, 18$, and $n = 3, 4, 5, 6, 7, 8, 9, 10$ respectively. Since our calculations are in good agreement with the available three experimental measurements, the other calculations provide reliable predictions for other ions. Tungsten ions contaminate the fusion plasma in thermonuclear reactor and dramatically influence the energy production. Therefore, our predictions are important for a plasma modelling of the fusion reactions.

Another manifestation of chaos is enhancement of the Raman photon scattering and suppression of the photoionization via chaotic compound resonances [4, 9].

We continue a general development of the statistical theory of finite chaotic quantum systems. Most recent result [4]: interference between chaotic compound resonances (neglected in independent resonance approximation) leads to a coherent contribution, which determines the energy-averaged total cross sections of electron- and photon-induced reactions. On the other hand, the partial cross sections (e.g., electron recombination, Raman photon scattering) are dominated by the stochastic contributions.

The statistical theory can be applied to intramolecular vibrational energy redistribution in polyatomic molecules, which is a key to a majority of chemical reactions.

References