Atomic Data Needs in Laboratory Astrophysics: Experimental Methods For Spectroscopy and Charge Exchange with Ions: A Summary

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I. SUMMARY

Modern astrophysics is reliant on ground- and space-based telescopes of increasing resolution and capabilities. Behind the scenes, however, are the decades of experimental studies used to interpret and understand all astronomical spectra. These experimental studies span a wide range from knowledge of calibration sources, to emission spectra used to interpret observed lines from astrophysical sources. Ultimately, the goal of all "laboratory astrophysics" pursuits is to reduce the uncertainties of atomic data such that uncertainties in astronomical observations and models are not determined by the underlying atomic data. When considering an astrophysical environment, the local conditions may give rise to a large number of charge states and species (atomic or molecular), each interacting via numerous physical processes, resulting in photon emission. Without knowledge of the wavelengths of emission lines, it is difficult or altogether impossible to attribute the emission to any particular species. Similarly, the competing physical processes e.g. electron impact or other collisional processes, each with their own probabilities and dependencies, ultimately contributes to the observed spectra. Disentangling this web of interactions and understanding the underlying physics requires experimentally-derived quantities such as cross sections (as functions of energy or other physical quantities), or physical models benchmarked by laboratory investigations. Without these laboratory data, the source (both species and mechanism(s)) of astrophysical emission cannot be determined with certainty.

In my dissertation, experimental studies were pursued in two critical areas: (1) understanding and measuring charge exchange cross sections for applications in x-ray astronomy, and (2) experimental investigations of open $5d$ shell spectra, such as that from neutral gold (Au), for benchmarking models of electronic structure and neutron star merger spectra known as kilonovae. To (1), gas targets (a gas cell and a gas jet) were developed to enhance the experimental capabilities of the Clemson University Electron Beam Ion Trap (CUEBIT) facility. In (2), the spectra of Au I and Au II were recorded and analyzed from a hybrid stellarator/Tokamak plasma apparatus at Auburn University. In the following Sections II and III, open questions in the two areas are discussed, and my contributions to the fields are summarized. Lastly, in Section IV, future applications and developments resulting from my work are discussed.

II. CHARGE EXCHANGE EXPERIMENTS

Charge exchange (CX) is the process by which an ion, A^{q+}, captures an electron from another species B (hereafter considered neutral), resulting in the population of a highly excited state, from which an electron cascade produces photon(s) γ. The net process may be written

$$A^{q+} + B \rightarrow A^{(q-j)+} + B^{j+} + \gamma \tag{1}$$

where j electrons are captured. CX with highly charged ions (HCI) typically results in x-ray emission. Given the highly charged nature of the ion (e.g. the 'projectile') and the neutrality of the 'target' B, this process is typically observed at the interface between hot plasma and cold neutral gas. Indeed, a common environment for this CX x-ray emission arises from the interaction between the solar wind and cometary comae [1]. CX emission is also observed in other exotic contexts such as supernovae remnants [2] or galaxy clusters and dense clouds [3]. These x-rays are highly diagnostic, arising from the unique x-ray signature of each ion, providing information not only of the emitting species, but also of the neutrals from which they are charge-exchanging. The cross sections for the process are very large (typically in order 10^{-15} cm^2 or larger), and vary with projectile charge state, collision energy, and even target species. The big question in CX x-ray emission is not *where* do these highly charged ions emit in wavelength, but *how*. Following population of a highly excited state, the resulting cascade depends on Einstein A coefficients, which for HCIs can be reliably calculated with any numbers of structure codes (such as e.g. The Flexible Atomic Code [4]), and are made available in places such as the NIST Atomic Spectra Database [5]. Instead, the big question is: *which states are populated following electron capture?*

While the principle quantum number (n) dependence is relatively well understood, the angular momentum (l) distribution of excited states populated by CX is much less understood. Various models are available for the l distributions of excited states

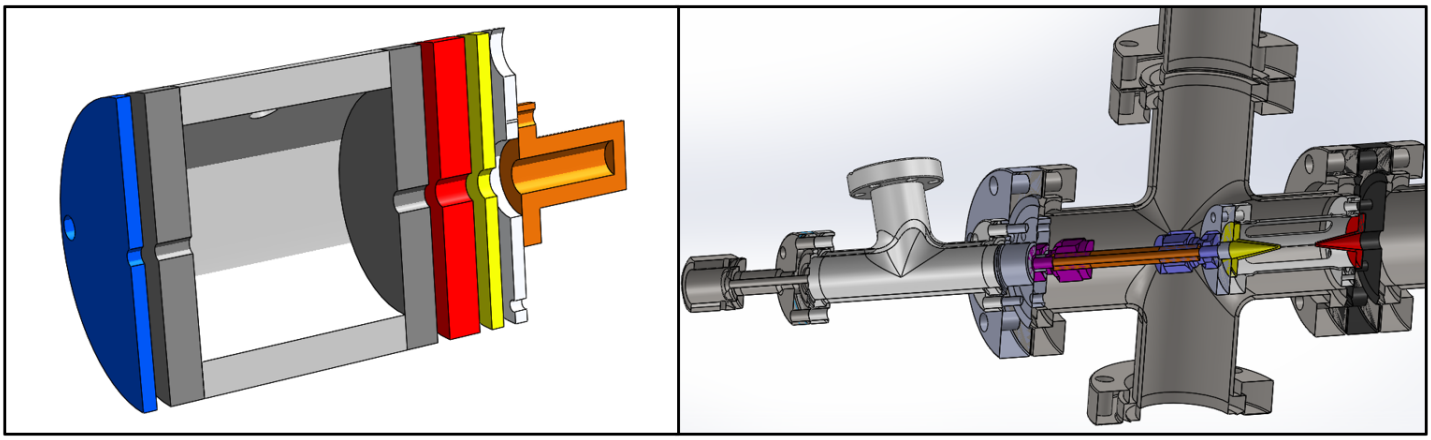

FIG. 1. Gas target schematics: (*Left*) Gas cell for measuring *total* charge exchange cross sections [10]. Gas is injected via a leak valve attached to the cell body by flexible stainless steel tubing, and pressure is measured by a hot ionization gauge via a separate but identical set of tubing. (*Right*) A low-pressure gas jet design for loading volatile organic compounds into CUEBIT.

(see e.g. [6, 7] and references within), but differences between modeled and observed spectra require experimental studies to benchmark and improve these models. While several excellent experimental works are available, such as the expansive work by Greenwood *et al.* [8], further work must be done. In some cases, such as $O^{6+} + H_2O$ [9], double electron capture is followed by autoionization. It is not yet clear in which cases bound double electron capture dominates over double capture autoionization. While models exist for single charge exchange and are expected to be benchmarked and improved in the coming years, double electron capture is still problematic. Theoretical efforts on double electron capture from multielectron targets are still being developed. Measurements of multielectron capture cross sections and accompanying x-ray lab spectra would greatly aid the development of such models.

In my work, two gas targets were developed for incorporation onto the Clemson University Electron Beam Ion Trap (CUEBIT) beamline to advance the study of CX x-ray emission. The two targets, shown in Fig. 1, are (1) a gas cell, used for measuring *absolute* total cross sections for both single- and multi-electron capture, and (2) a gas jet for enabling the production of metal ion beams from CUEBIT.

The gas cell is capable of measuring absolute, total CX cross sections by measuring attenuation of an ion beam as a function of target pressure. The CX cross section, σ_{cx}, may be written in terms of measurable quantities via [10]:

$$\sigma_{cx} = \frac{k_b T}{PL}\left(\frac{I_o - I}{I_o}\right) \tag{2}$$

where k_b is the Boltzman constant, T and P are the temperature and pressure of the target gas, L is the path length of the ion-gas interaction region, and I_o and I are pre- and post-gas beam currents. By measuring an initial beam current I and the beam current with gas, I_o, the CX cross section can be directly calculated. However, this method is not so straightforward when multielectron capture contributes. In addition to enabling the measurement of ion beam energy [11], the use of a retarding field analyzer [10] allows for measurement of beam currents in each charge state, from which the cross section of multi-electron capture can then be calculated. In preparation of future uses with highly charged ion beams, the gas cell was benchmarked by measuring well-known CX cross sections of singly charged noble gas ions [12], and subsequent exploratory measurements of CX between single charged ions and molecular nitrogen were reported in [13].

The second gas target, the gas jet (Fig. 1), was developed for enabling the production of astrophysically relevant metal ions such as Fe^{16+} at the CUEBIT facility. The gas jet consists of a nozzle and skimmer system, machined using standard tooling and constructed inside commercial vacuum hardware, which can be pressurized and operated with any gas. Attachment to a viewing port of CUEBIT and backing with a high pressure results in a ballistic flow along the direct line-of-sight between the skimmer exit and CUEBIT drift tubes. The jet could replace the existing gas injection scheme for producing e.g. Ne or Ar ion beams. Alternatively, the nozzle system could be pressurized by a heated volatile organic compound (VOC) source for producing metal (Mg, Fe, ...) ion beams from easily-sublimable metal-containing compounds. This technique, "Metal Ions from Volatile Organic Compounds" (MIVOC) has been utilized at other beamlines (e.g. [14]) but has not yet been attempted at CUEBIT. The large nozzle size (diameter ~ 0.014") ensures sufficient target density while minimizing the risk of clogging and reduces the demands of differential pumping. Initial tests of the nozzle system, pressurized with N_2 gas, were conducted

and target densities in a test setup agreed reasonably well with a simple thermodynamic model from Ref. [15]. Discrepancies in the inferred and expected target densities were attributed to large uncertainties in conductances and pumping speeds in the test apparatus.

III. GOLD SPECTROSCOPY

As reflected in the number of citations and papers in the NIST database, the spectra of row 6 elements with open $5d$ shells, save for tungsten, have received relatively little interest in the past few decades. With the recent detection of a neutron star merger (NSM) by the LIGO collaboration [16], low charge states of row 6 elements such as the 3rd r-process peak (Au, Pt, ...) have received increasing interest. While studies of their importance and contribution to the opacity of neutron star merger observations are available (e.g. [17, 18] to name a few), few laboratory experiments exist to benchmark the structure calculations underpinning NSM models. The required atomic data spans a large range from the lanthanides and actinides to relatively simpler elements such as gold or platinum.

On their own, these elements are particularly interesting to atomic physicists. The elements are heavy, with masses in excess of 150 Da. As a result, fine structure splittings are significant; for example, the splitting of the ground state of neutral Fe is of order 10 meV, but similar splittings in row 6 elements such as Pt may be of order 0.5 eV or more! The open $5d$ shell, when coupled to excited $6s$ or other electrons, leads to a large number of levels complicated by strong mixing. Calculating the electronic structures accurately requires a detailed treatment, including core-valence coupling, core polarization, configuration interaction, and additional relativistic effects [19]. The difficulty in calculating the electronic structure of these elements is expanded on by Ref. [20], who mentions that no general-purpose code is yet available to generate data at or near spectroscopic accuracy for these elements. Development and benchmarking of new structure codes would greatly benefit from experimental characterization of these systems.

In an effort to expand on the experimental literature of these elements, I had undertaken a study of neutral and ionized gold. Historically, spectrosopic surveys of these elements utilized hollow cathode discharges, from which high-resolution Fourier transform or grating spectroscopy from the UV and visible ranges yielded signficant insights into their structures. Early works on Au I and Au II by Platt & Sawyer (1941) [21] and Ehrhardt & Davis (1971) [22] revealed much of the Au I structure, including a number of doubly excited levels and ns, np, and nd Rydberg series. Later, Rosberg & Wyart observed Au II in the vacuum ultraviolet, and reported accurate wavelengths and transition probabilities from judicious applications of Cowan's suite of codes [23].

In my studies, the spectra of Au I and Au II were observed inside the Compact Toroidal Hybrid (CTH) plasma apparatus at Auburn University [24]. Spectra were collected between 187 - 800 nm from dozens of discharges with a gold-plated probe inserted at various depths inside the plasma. To aid in line identification, spectra were also collected with a nickel-plated probe, which was invaluable for separating out contaminant lines from those of gold charge states. The higher temperatures in the CTH ($T_\mathrm{e} > 10$ eV) are far in excess of those in hollow cathode discharges, and excitation tends toward more highly excited levels than those reported in previous studies. Though no new levels were reported in this work, lines belonging to the most highly excited known configurations of Au I ($5d^9 6s6d$ autoionizing levels) and Au II ($5d^8 6s6p$) were found in abundance.

Grotrian diagrams of Au I and Au II are shown in Fig. 2. Known levels are indicated by horizontal lines above their configuration labels; previously-known lines which were also found in our spectra are shown in blue, with newly observed lines shown in red. The results are reported in Bromley *et al.* [25], totaling 86 lines of Au I, 43 of which were previously unobserved, and 76 lines of Au II, 51 of which were previously unobserved. Interestingly, many of these transitions are "two-electron" transitions, whose detection is enabled by the significant configuration mixing and breakdown of LS selection rules in heavy elements. While hundreds more emission lines of Au I and Au II are known from previous works, many could not be detected given impurities in the plasma arising from the probe design (e.g. hundreds of line from Fe I and Fe II), and limited intensity response compared to the photographic plates of previous works. Further analysis of the gold spectra and applications of our data will be discussed below.

IV. FUTURE APPLICATIONS AND IMPLICATIONS

Future telescopes, such as *Xrism* or *Athena*, are anticipated to launch with high spectral resolution which will increase the demands for laboratory astrophysics. My work had focused on developing and using experimental techniques for measuring fundamental atomic properties useful to astrophysicists. However, one can imagine several further investigations stemming from my work. In the arena of charge exchange x-ray emission, the gas jet, backed by volatile organic compounds, may enable the CUEBIT beamline to produce beams of highly charged metals such as Mg^{12+} relevant to cometary studies. These HCI

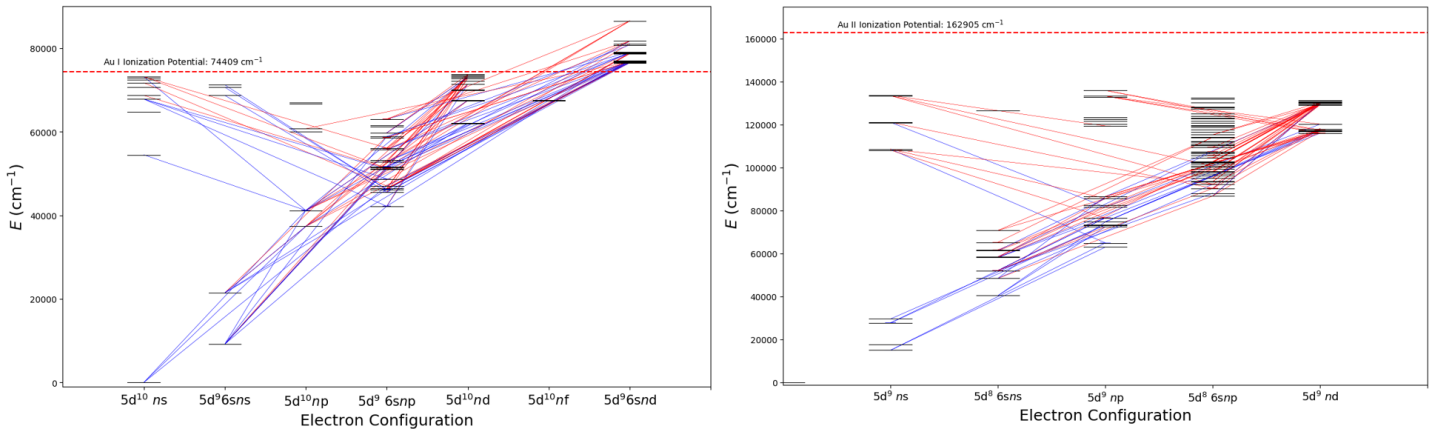

FIG. 2. Grotrian diagrams of Au I (left) and Au II (right). All known doubly excited levels through $n, n' = 6$ are shown. In Au I, Rydberg series up to $n = 14$ are shown when possible. Previously known transitions also observed in our spectra are indicated in blue, with newly observed transitions shown in red. Ionization limits are shown as dashed horizontal lines.

beams will enable further studies of CX x-ray emission in crossed beam experiments at the end of the CUEBIT beamline. As part of my present postdoc, a Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) apparatus is being designed and constructed at Auburn University prior to relocation to the CUEBIT facility. Such an experiment will yield x-ray spectra and direct measurements of n distributions following CX, from which the nl distribution of capture states may be inferred. Additionally, a new method recently developed by Betancourt-Martinez *et al.* [26] to extract state-selective cross sections from x-ray spectra would simplify this analysis considerably. However, additional work will be required to properly account for missing forbidden line emission which is commonplace in astrophysical spectra but absent from laboratory spectra given the small forbidden transition rates. Where/if necessary, the gas cell I have constructed and tested will enable total, absolute cross section measurements at the CUEBIT facility which can be used to place state-selective cross sections onto an absolute scale. While not directly beneficial for the analysis of individual emission lines, it would enable refinement of ionization balance calculations which can affect line intensities within spectral models. In total, the gas targets developed in my work prepared the CUEBIT facility to conduct any number of studies aimed at revealing the diagnostic potential of charge exchange x-ray emission in astrophysical plasmas.

On the other end of my work, the gold spectroscopy carried out thus far is the first of its kind. My work showed the validity of using a plated target inside a tokamak for line identification purposes, and suggests future experiments with solid targets of other row 6 elements (e.g. platinum) will yield similar excitation of the more highly excited and difficult-to-access configurations. One can imagine several areas of investigation involving the collected gold and nickel spectra. First, the nickel data contains a number of as-yet-unidentified lines which are decidely not gold, and a detailed analysis of the nickel spectra is being conducted by students at Clemson University. It is expected that just as in gold, the more highly excited levels of Ni I and Ni II will be accessed, providing new identifications for astronomical spectroscopy.

Second, the gold spectra were collected from a plasma in non-LTE conditions, and as such would be invaluable for benchmarking collisional datasets over a wide range of temperatures and densities. The usefulness of these spectra are shown in the recent study of W I by Smyth *et al.* [27], who used CTH tungsten spectra to benchmark collisional data for neutral tungsten in fusion plasmas. It is expected that the metastable levels of the Au I $5d^9 6s^2$ configuration, similarly to what was found in the recent Johnson *et al.* study of W I metastables [28], will contribute strongly to the population dynamics. One can imagine a similar benchmarking process being carried out for gold collisional data when/if it is available. Additionally, our spectra may be used to identify nickel or gold emission in astrophysical plasmas directly. Given the small number of contaminants (C, N, O, Fe, ...) in our spectra, comparison of our spectra to those from astrophysical sources sharing few contaminants may directly reveal the positions of as-yet-unidentified emission lines in astrophysical spectra. While forbidden transitions from the metastable $5d^n 6s^2 (n = 9, 8$, respectively) configurations of Au I and Au II to ground were not observed here, the recent work in W I by Ref. [28] suggests they may be observed in astrophysical environments if densities are low enough to enable a significant population of metastables.

Lastly, the digital format of the spectra allows for future re-analysis when structure calculations of these systems become available. In Au I, the configurations and levels are known, but A values are only available for a select few lines. My spectra may be useful for placing uncertainties on A values for lines from the same upper levels. In Au II, no levels between ~135,000 cm^{-1} and the ionization limit are known; comparisons to isoelectronic Pt I suggests these are the undiscovered $5d^9 5f$ or

$5d^8 6s6d$ configurations, which suffer from signficant mixing. When structure code results become available, comparison between calculated line intensities and the gold spectra may lead to the identification of new levels. To benefit future calculations of other $5d$ elements such as platinum or iridium, solid targets are being developed for spectroscopic measurements inside the CTH. Spectra from these probes are expected to act as benchmarks for the electronic structure calculations being carried out by colleagues at University of Georgia and Queen's University Belfast.

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