From Molecular to Highly-Charged Ions: Expansion of Laboratory Astrophysics Through Use of the Electrostatic Storage Ring with Electron Beam Ion Trap and Electrospray Ion Sources

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ABSTRACT

Described herein is the use of an electrostatic storage ring (ESR) coupled to a variety of ion sources to generate and store positive and negative molecular ions, as well as highly-charged positive ions. Studies of collision phenomena within each class of ion targets provide insight on plasma properties of astrophysical objects. These objects range from cool molecular clouds – detected through their infrared absorption and emission spectra – to stellar mass ejections and quasars with highly-charged ion (HCI) emissions – detected by their X-ray emission spectra. A summary is given of the capabilities attained by combining an ESR with molecular-ion sources, or an electron beam ion trap for HCIs. This work anticipates the continued rich return of space observations from, for example, Spitzer, SOFIA, Herschel, and ALMA in the infrared region; to SOHO, Chandra, XMM-Newton, Constellation X and NEXT in the EUV and X-ray regions. The ESR enables one to measure molecular phenomena such as direct and dissociative ionization, direct and dissociative recombination, and lifetimes of negative ions. For HCIs, one can study direct and indirect ionization, direct and dielectronic recombination, excitation, and lifetimes of levels in the 10^{-9} to 10^{-2} second range. Because there is no magnetic field, Zeeman mixing of excited levels is obviated; and since the ions circulate for ≈10-100 seconds, the targets are in their ground vibrational-electronic state.

INTRODUCTION

The astounding advances in astrophysics through ground-based spectrometer observations, and through NASA's and ESA's flight spectrometers, have extended our view of the Universe from the far infrared to the X-ray region of the electromagnetic spectrum. Rich molecular absorption and emission spectra are observed by the National Radio Astronomy Observatory, ISO, NICMOS, and Spitzer; with measurements to be expanded by the upcoming SOFIA and Herschel missions. Spectra of highly-charged ions (HCIs) are observed in our Sun, stars, and quasars by EUVE, Lyman/FUSE, Suzaku, SOHO, Chandra, and XMM-Newton. Measurements will be expanded by the upcoming Constellation X and NEXT missions. Examples of molecular infrared spectra observed by Spitzer are given in Fig. 1 [1,2]; and the rich HCI emissions from Capella observed by Chandra and XMM-Newton are shown in Fig. 2 [3].

Underlying these measurements is the need for a broad understanding of the plasma properties of the various astrophysics objects. Such understanding is intimately connected to the underlying atomic and molecular collision physics. For molecular-ion targets, there is an ongoing need for accurate cross sections and rate constants for direct and dissociative ionization,

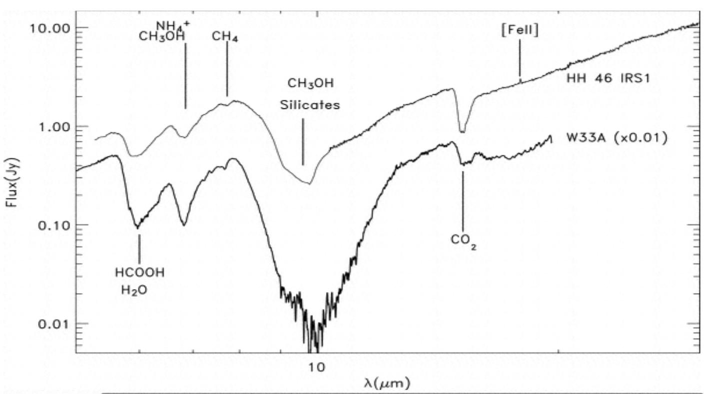


Figure 1a: ***Spitzer*** **IRS ionic and molecular spectrum of the HH 46 IRS 1 source (*upper line*) compared with that of W33A (*lower line*), a high-mass protostar, scaled down by a factor of 100 (from Refs. [1,2]).**

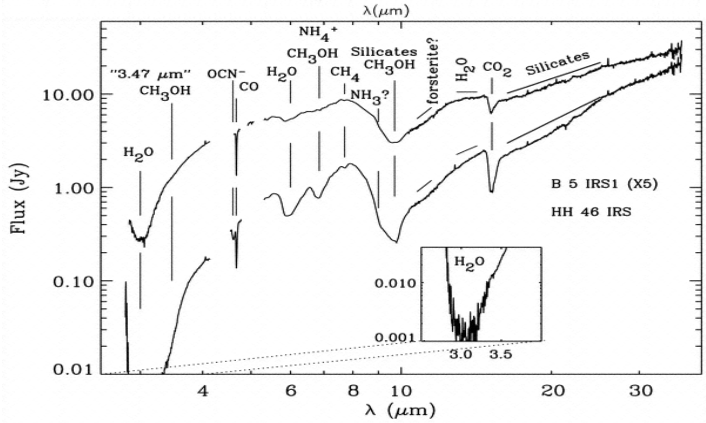


Figure 1b: Combined *Spitzer* and ground-based *L*- and *M*-band spectroscopy of B5 IRS 1 (*top*; multiplied by factor of 5 for clarity) and HH 46 IRS (*bottom*) (from Refs. [1,2]).

direct and dissociative recombination, and electron detachment. For HCIs one requires cross sections for excitation, direct and dielectronic recombination, single- and multiple-charge exchange (in HCI-neutral collisions), HCI level lifetimes (nanosecond to millisecond range), and *f*-values. A review of these processes and needs may be found in Refs. [4,5].

Since one cannot measure every required cross section or lifetime, benchmarking of theoretical results is a critical and important aspect of laboratory astrophysics. As an example, recent JPL results of absolute e-Fe^{13+} excitation cross sections have helped resolve the so-called "Iron Conundrum," in which the under-abundance of the Fe density (relative to Mg, Si, Ca, *etc.*)

in Seyfert Galaxies compared to its density in our Sun was successfully explained by a too-large collision strength for the Fe^{13+} $^2P^o_{1/2} \rightarrow ^2P^o_{3/2}$ fine-structure transition calculated in an 18-State R-Matrix theory, relative to that of the more accurate 135-State Breit-Pauli R-Matrix theory [6]. While the topic is not addressed herein, the development of accurate (10% level) theoretical approaches to the calculation of astrophysical phenomena is also essential to meeting NASAs and ESAs ongoing and future, high-quality space observations.

EXPANDING THE CAPABILITIES FOR LABORATORY ASTROPHYSICS

The profound extension of space observations into the infrared and X-ray regions of the spectrum has revealed a rich population of new molecules and highly-charged ions! In order to address the current and anticipated rich return of space data, one now requires a concomitant expansion of experimental methods and facilities for measuring (in *molecules*) ionization and recombination cross sections; and (in HCIs) collision strengths, lifetimes, charge-exchange cross sections, ionization cross sections, direct and dielectronic recombination cross sections, and

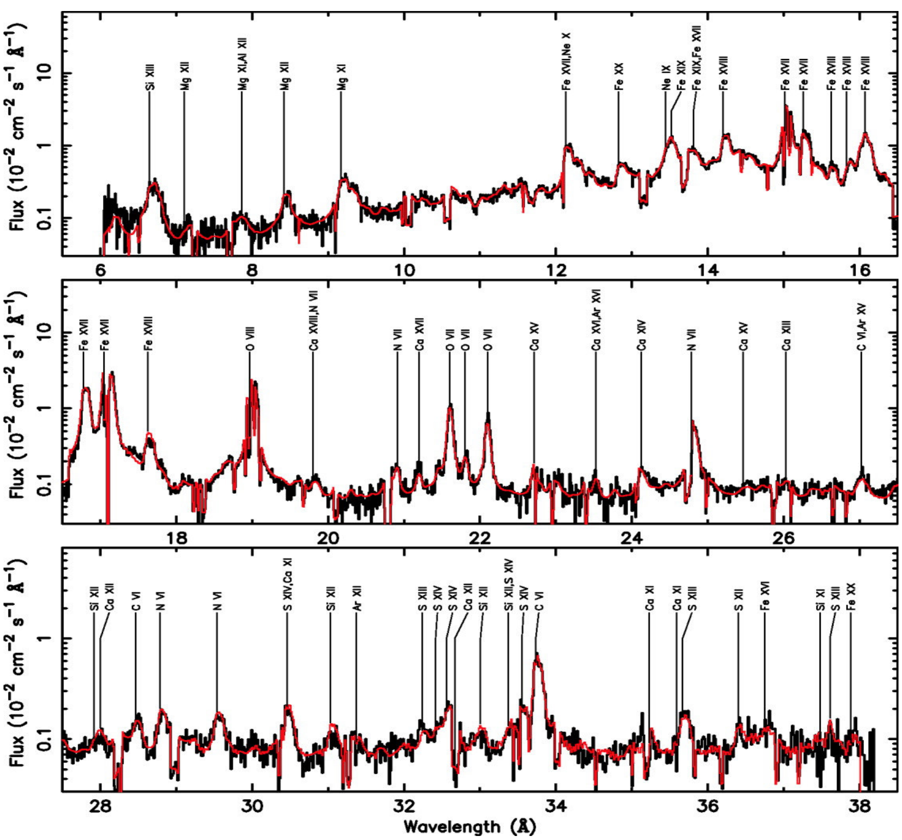


Figure 2: X-ray spectra of the rich HCI emission features at the Capella corona observed by the *Chandra* and *Newton* spectrometers.

Comparison is given of the measured (black) and simulated (red) emission features [3] Excitation of the HCI emissions results from hot electrons in the stellar corona.

lifetimes. Many of these phenomena (cross sections and lifetimes) are already being measured. Use is made of the electron energy-loss method (developed at JPL) with merged beams of electrons and HCIs [6]. Single and multiple charge-exchange cross sections are measured using a collision gas cell and retarding potential difference analysis of the charge-exchanged HCI ions [7]; and metastable lifetimes are measured using a Kingdon ion trap [8]. A photograph of the JPL facility is shown in Fig. 3, and a close-up of the charge-exchange beam line is shown in Fig. 4. Three separate beam lines are involved. Provided in Figures 5a-5c are results from each of the three beam lines. Shown in Fig. 5a are absolute experimental and theoretical excitation cross

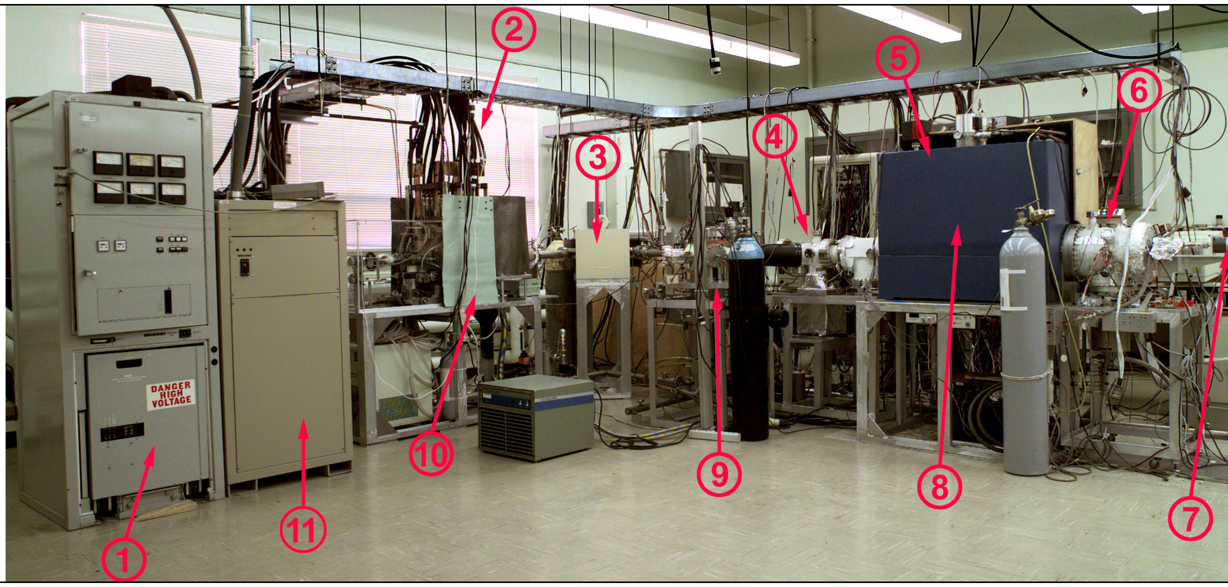


Figure 3: The JPL Highly-Charged Ion Facility. The numbered sections are, ① high-power Ku band amplifier, ② cooling lines for the *Caprice* HCI source solenoidal magnets, ③ HCI charge/mass selection magnet, ④ Y-switcher for directing the HCI beam into one of three beam lines, ⑤ solenoidal magnet for merging electron and HCI beams, ⑥ vacuum manifold for electrical feedthroughs, ⑦ stepper motor for measuring beams profiles, ⑧ merged-beams chamber for measuring absolute excitation cross sections, ⑨ Kingdon ion trap for measuring HCI lifetimes, ⑩ *Caprice*-type HCI source with lead shielding, **(11)** 1000-A supply for the *Caprice* solenoidal magnets.

sections for the Fe^{13+} coronal green line, by which the so-called "Iron Conundrum" was resolved [6]. Shown in Fig. 5b is the *lifetime* of the coronal green line transition, as measured in the Kingdon ion trap beam line [8]. And shown in Fig. 5c are the modeling results of Ref. [9] for X-ray emission from the comet McNaught-Hartley, in which published JPL absolute charge-exchange cross sections were used for solar wind-comet neutral collisions.

There are distinct enhancements to the present JPL approach. For example, **(1)** the *Caprice*-type electron cyclotron resonance ion source can produce charge states in Fe only to about Fe^{15+}, whereas one requires beams of Fe^{18+}-Fe^{25+} ions for measuring cross sections applicable to energetic solar-flare emissions and coronal mass ejections. **(2)** In some cases, one requires a better measure of the metastable content of an HCI beam; or better yet, one would like

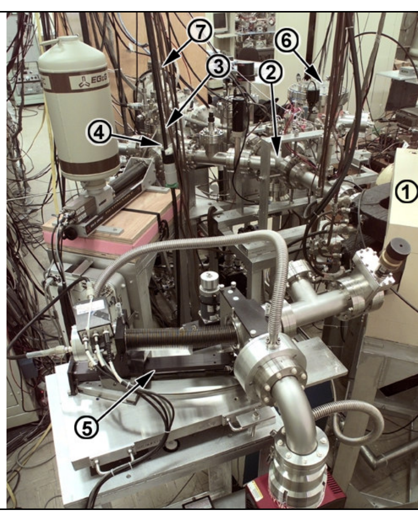


Figure 4: Close-up of the JPL Charge-Exchange Beam Line. The numbered sections are, ① HCI charge/mass selection magnet, ② Y-switcher for direction the HCI beam into one of three beam lines, ③ charge-exchange gas cell, ④ HPGe X-ray spectrometer, ⑤ U. Connecticut X-ray grating spectrometer (here, not attached to beam line), ⑥ Kingdon ion trap for measuring lifetimes, and ⑦ merged-beams chamber for measuring excitation cross sections.

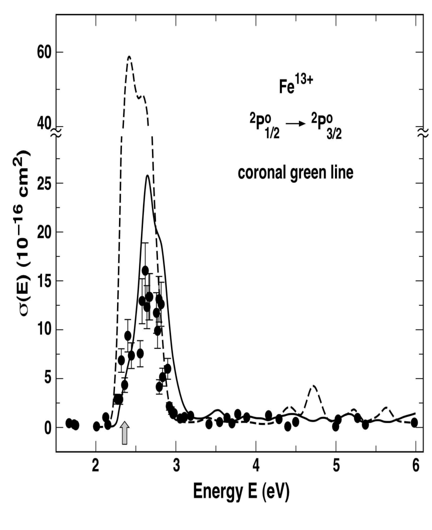


Figure 5a: The "Iron Conundrum" resolved: comparison of JPL experiments (●) and an accurate theory (—) for the Fe^{13+} coronal green line cross section [6]. A less-accurate theory (----) led to a too-low Fe stellar abundance.

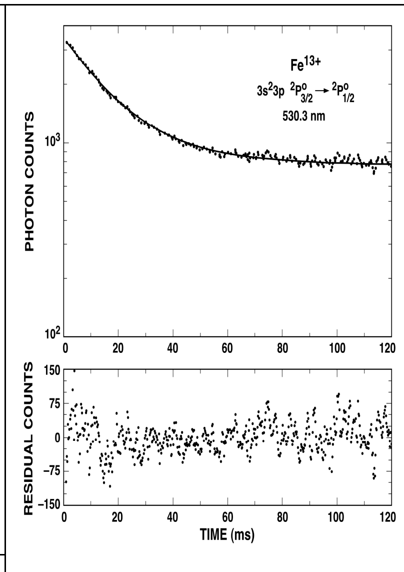


Figure 5b: Photon decay rate for the Fe^{13+} coronal green-line transition $3s^2 3p^2$ $^2P^o_{3/2} \rightarrow {}^2P^o_{1/2}$ at 530.3 nm [8]. The measured lifetime is 17.0±0.2 ms.

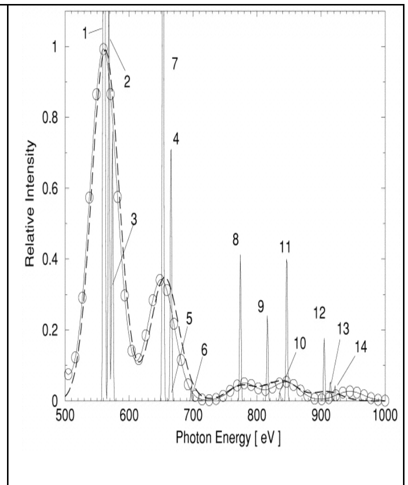


Figure 5c: Comparison of observation (o) and fit (----) to the X-ray emission spectrum from Comet McNaught-Hartley using JPL charge-exchange cross sections [9].

to have a *cool* beam where all metastable levels have decayed. A translational energy spectrometer is being designed to quantify the metastable content in the JPL beams, but this is a somewhat unwieldy subsystem requiring beam line changes and additional power modules. **(3)** One would like to have a means of measuring lifetimes in the 10^{-9}-10^{-2} range in just a single experimental apparatus. **(4)** To measure collision phenomena in molecular ions, one requires a means of changing ion sources to generate negative ions (use a hollow-cathode glow discharge), positive ions (use a Nielsen-type source), or large clusters and biomolecules (use electrospray ionization). **(5)** The experimental system should have a straightforward means of studying direct and indirect electron ionization and dissociation phenomena for both molecular and HCI targets. A holistic approach to the issues in (1)-(5) above is to design and build an electrostatic storage ring (ESR), coupled to an electron beam ion trap. A photograph and schematic of the tabletop ELISA (**E**lectrostatic **I**on **S**torage **R**ing, **A**arhus) are given in Figs. 6a and 6b, respectively. ELISA was originally built at Aarhus University in Denmark [10-12]. It has served as the model for rings at the Tokyo Metropolitan University, and at the KEK, Tsukuba. Coupling the ESR to a variety of ion sources would allow one to access an extremely broad range of molecular and HCI targets. Molecular ions of astrophysical interest include CH^+, H_3^+, H_3O^+, CN^-, CO^-, and C_4H^-; and HCIs include $Mg^{8\text{-}11+}$, $Si^{5\text{-}13+}$, $Ca^{10\text{-}19+}$, and $Fe^{15\text{-}23+}$. Large molecules, clusters, and biomolecules will also be injected into the ESR using electrospray (ES) ionization methods at Aarhus [11], or micromachined sources being developed in collaboration with JPL's MicroDevices Laboratory (P. Willis, private communication 2009).

Generation of HCIs is considered a critical part of understanding solar and stellar spectra, including solar and stellar winds interacting with neutral clouds and comet atmospheres to generate X-rays (see, for example, Refs. [7,9] and citations therein). As such, an integral part of the infrastructure to laboratory astrophysics will be the addition of a state-of-the-art HCI source.

Figure 6a: Photograph of the ELISA electrostatic storage ring at the University of Aarhus [10,11]. In addition, a double (merged) ESR ring system is in the final phases of construction at Stockholm University [13]. The dimensions of the ring are 2m (length) × 1m (width).

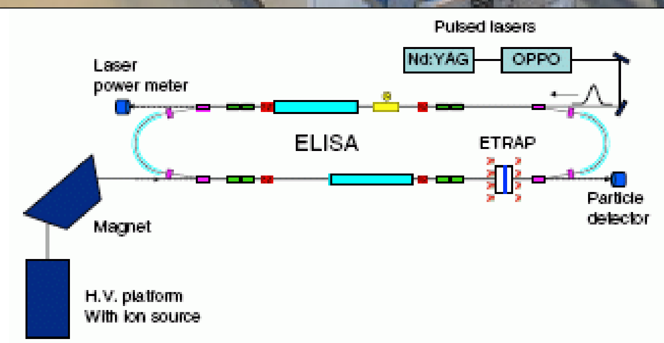


Figure 6b: Schematic Diagram of the Aarhus ESR. System is shown with an Nd:YAG laser, pumped tunable optical parametric oscillator (OPPO), and electron- gun target (ETRAP). It is configured to measure dissociation of the target molecular ions using the array particle detector [10,11].

The Refrigerated Electron Beam Ion Trap (REBIT) can generate practically any charge state of any ion, and will provide coverage for the vast majority of HCIs encountered in astrophysics. A schematic diagram of the REBIT is shown in Fig. 7a, together with a mass spectrum of xenon charge states up to Xe^{45+} displayed in Fig. 7b. The source is commercially available, and would be procured as part of the system build [14].

NEEDED LABORATORY DATA AND RECOMMENDATIONS

The needs for laboratory data for understanding the plasma environment of solar, stellar, and interstellar objects are vast. In addition to providing cross sections to the modeling community accurate, measured absolute cross sections are critical to benchmarking results of calculations. Not all cross sections, lifetimes, branching fractions, collision rates, *etc.* can be measured, and hence the most accurate theories must be used to calculate missing data.

The combination of the ESR with the REBIT (HCI) and ES (molecular anions and cations) sources will provide a veritable universe of experimental capabilities. Some of these classes of experiments are given in Table 1. It is clear that a properly-configured experimental atomic and molecular physics laboratory will of essence provide experimental data that encompass a broad range of astrophysical applications and missions. The ESR and REBIT combination described herein will provide measurement capabilities in which there is no "wavelength barrier" or "ion barrier"! Using the JPL electron energy-loss technique [4,6] transitions from the infrared to the hard X-ray regions are accessed. Since the ESR is completely electrostatic,

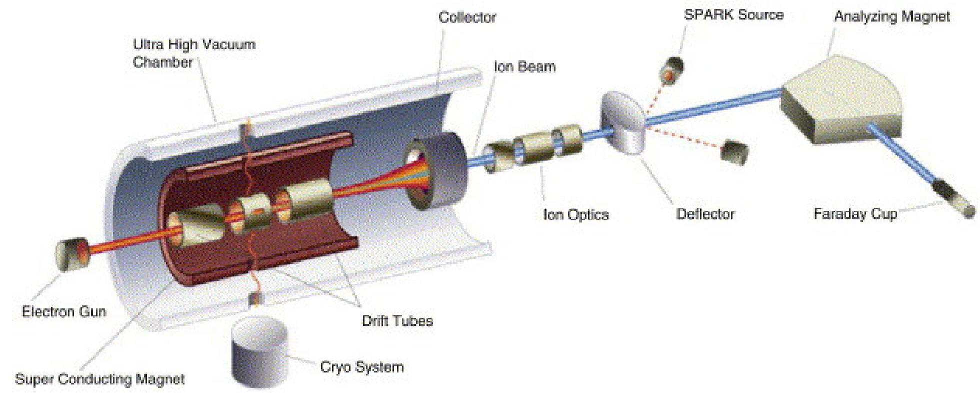


Figure 7a: Schematic of the Refrigerated Electron Beam Ion Trap (REBIT). Practically any charge state of any ion can be generated, with ample particle current for use in the ESR [14].

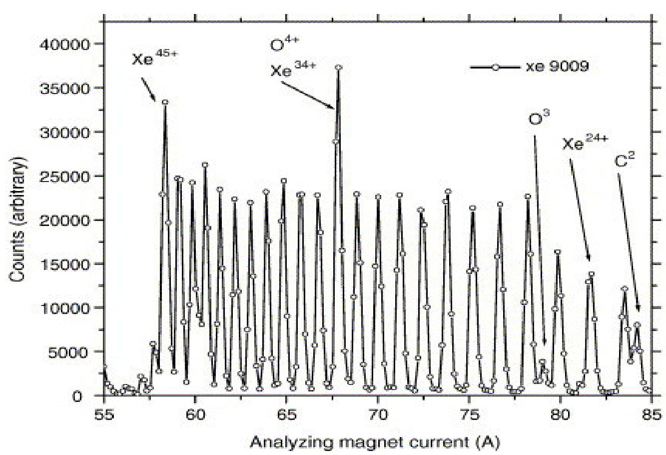


Figure 7b: Ion Charge States Using the REBIT. Production of fully-stripped Fe ions is also possible. The generated ions and charge states will cover an extremely broad range of ions presently observed and anticipated in astrophysical objects [14].

trapping depends only on target energy and *not* mass: species from hydrogen-like HCIs, to diatomic molecules, to DNA will be generated and trapped!

We also point out that the needs of laboratory astrophysics described herein for furthering NASA, NSF, and DoE space observations resonate with a recent report by the AAS Working Group on Laboratory Astrophysics [15]. One recommendation of the AAS study is to nurture instrumentation, technology, and facilities development programs in laboratory astrophysics for development, construction, and maintenance of state-of-the-art laboratory astrophysics instrumentation and facilities.

COST AND SCHEDULE

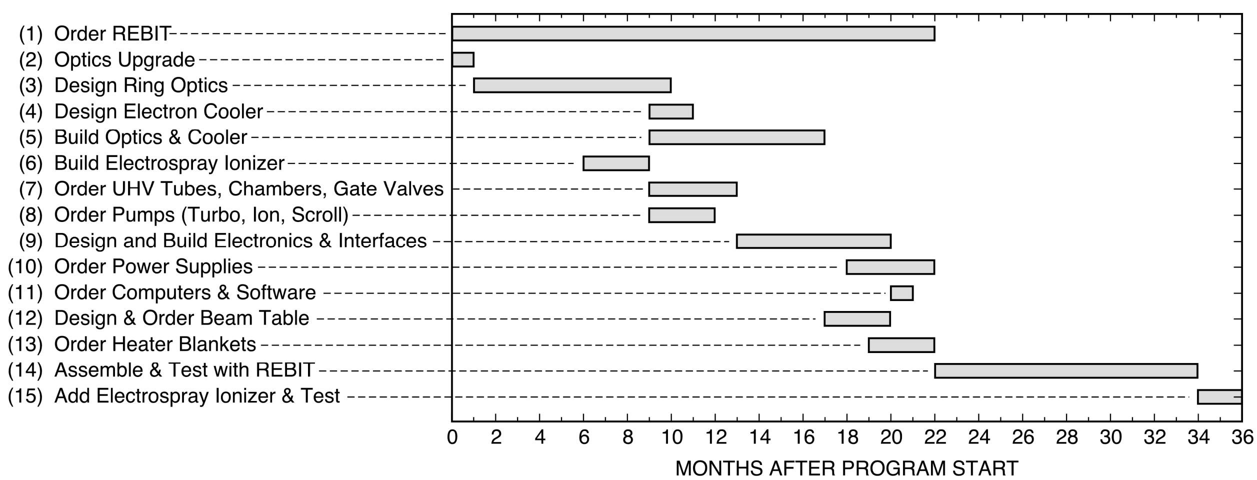
Design, fabrication, assembly and test of the system are estimated at $5.3M (FY09 dollars) to cover 36 months. A milestone schedule is given in Table 2. An estimated budget for the ESR-REBIT-ES facility in terms of combined, burdened, labor and procurements in each of the three years is as follows: **Year 1**: $2.7M, **Year 2**: $1.5M, and **Year 3**: $1.1M. Included in Year 1 is travel for personnel between JPL and Aarhus (ELISA) and Stockholm (DESIREE) [13] to confer and to broaden collaborations.

There are three ESRs in the world: two in Japan, and the original ring at the University of Aarhus, Denmark. The system described herein would be the first ESR in the United States. It would both sharpen and continue the JPL focus on understanding astrophysical processes in the laboratory. The ESR facility at JPL will add to the *scientific infrastructure* of the laboratory. The connection between good physicists at JPL and the success of flight projects is subtle.

Table 1. Summary of the Experimental Capabilities in Molecular and HCI Collisions Enabled by the ESR-REBIT-ES System

• measurements of absolute electron-impact cross sections at energies of threshold to 5× threshold for up to helium- and hydrogen-like HCIs • expanded capability for understanding heavy ion ejection in supernovae, burst spectra in neutron stars, hot stellar plasmas during nucleosynthesis, *etc.*	• measurements of lifetimes in the 10^{-9} to 10^{-2} sec range for excited states of HCIs • expanded capability to the present approach (Kingdon trap, instrument geometry) limiting us to lifetimes of 0.5-5 ms
• absolute single and multiple charge-exchange cross sections for the ring-cooled, ground-state HCIs • expanded capability: our present approach uses more cumbersome and less accurate methods of measuring metastable populations by gas attenuation, and by translational energy spectroscopy (planned)	• measurement of negative-ion lifetimes in diatomic and polyatomic negative ions, in the absence of magnetic-field mixing and black-body (photodetaching) radiation • new capability for measuring molecular ion lifetimes dissociation pathways, in simple diatomic to large biomolecules
• absolute direct and indirect ionization cross sections of HCIs • new capability by crossing the ring ions with an electron beam to measure absolute single and multiple ionization phenomena	• absolute direct and dielectronic recombination cross sections in HCIs • new capability for understanding collisional ionization equilibrium in the Sun and stars by use of a merged or inclined electron beam on the stored HCIs
• accurate measurements of HCI fluorescence lifetimes (10^{-9}-10^{-2} sec) and isotope ratios by tunable laser absorption • for measuring isotope ratios, a new capability that overlaps our high-resolution Paul ion trap program	• laser photodissociation with product distributions of prevalent ISM ions such as H_2^+, H_3O^+, CH_3^+, CN^-, CO^-, and C_4H^- • new capability enabled by the electrospray (ES) source and the ESR geometry

Table 2. Milestone Chart for the ESR with REBIT and ES Ion Sources



Physicists are attracted by challenging research. In our experience, the collision physics research proposed here will attract scientists who will *also* become significant contributors to JPL flight projects. We also believe that the ESR in the United States will add to this *nation's* scientific infrastructure. The research team represents renowned colleagues in experimental negative- and positive-ion molecular chemistry (Adams), theoretical and computational quantum chemistry (McKoy), charged-particle optics design (Chutjian, MacAskill, Madzunkov), ion traps (MacAskill, Madzunkov, Chutjian), use of HCIs in experiments – including ion sources and ultrahigh vacuum techniques – (Simcic, Chutjian, MacAskill, Madzunkov); together with experts at the DESIREE ring in Stockholm (Schuch). This list of co-workers will almost certainly expand as we hire one or more NASA Postdoctoral Fellows, and as we engage the Japanese and European groups more fully.

The JPL Atomic and Molecular Group has had over the past seven months detailed discussions of the intent and ideas herein with NASA sponsors in the three program offices that presently support our HCI research at JPL/Caltech. (As a courtesy, this document has also been forwarded to the NASA program managers.) It is clear from these discussions that the capabilities listed in Table 1 are congruent with the missions of NASA's Laboratory Astrophysics, Planetary Atmospheres (including comets), and Solar and Heliospheric Physics Program Offices.

ACKNOWLEDGMENT

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