TITLE: EBIT in the Magnetic Trapping Mode: Mass Spectrometry, Atomic Lifetime Measurements, and Charge Transfer Reactions of Highly Charged Atomic Ions

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Mass Spectrometry, Atomic Lifetime Measurements, and Charge Transfer Reactions of Highly Charged Atomic Ions

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Abstract. Although it may sound like a contradiction in terms, the electron beam ion trap (EBIT) works as an ion trap even when the electron beam is switched off. We present various experiments that exploit the „magnetic trapping mode“ for investigations of ion confinement, charge exchange processes, atomic lifetime and ion mass measurements.

INTRODUCTION

Since its introduction, the electron beam ion trap (EBIT) [1] has played a significant role in the field of atomic x-ray spectroscopy. Similar to the electron beam ion source (EBIS), production and storage of highly charged atoms are effected by the use of an intense electron beam (typically of the order of a hundred milliamperes) of high energy (a few to a few hundred kiloelectronvolts) which is compressed by a strong magnetic field (several Teslas). The electron density in the beam is in the $10^{12}$ cm$^{-3}$ range thus exceeding the rest gas density of typically $10^6$ cm$^{-3}$. In contrast to the conventional EBIS configuration, the magnetic field is provided by a set of Helmholtz coils which allows x-ray and optical windows for radial line-of-sight access to the center of the trap. The pioneering work at Livermore has been followed by several research groups all over the world.

Conventionally the x-ray production is closely related to the interaction of the stored ions with the electron beam, e.g. by electron impact excitation, by radiative or dielectronic recombination. Thus, it is not obvious what could be gained from
switching off the electron beam once the highly charged ions are created. Since the middle of the nineties, however, a series of such experiments has been performed that aimed at a mass analysis of the trapped ion ensemble [2-4]. To this end, the two electron beam ion traps at Livermore, SuperEBIT and EBIT-II, were operated in a Penning-trap-like „magnetic trapping mode“ (MTM), in contrast to the „electron trapping mode“ (ETM) during ion production [5,6]. In the new mode, the drift tube voltages provide an axial electric trapping potential that complements the Lorentz force for radial ion confinement.

When the electron beam is switched off, the plasma changes within a fraction of a millisecond from being electron-dominated to ion-dominated. The space charge compensation provided for the ion cloud then ends as well as the attractive potential. Consequently, the ion cloud expands somewhat, as was recently quantified by the NIST EBIT group [7]. Distant ion-ion collisions redistribute the angular momenta with respect to the trap axis. Work at LLNL found this process to be irreversible: when the electron beam is switched on again, even as soon as after 1 ms, the ion cloud does not shrink back to its old size, but a new (electron beam-dominated) cloud is being built up from neutral atoms and low-charge ions along the axis, i.e. inside of the old ion cloud. The actual transition between the two steady-state conditions may become the subject of future ion-cloud dynamics studies.

**MASS SPECTROMETRY**

For a mass spectrometric (MS) analysis of the ion ensemble, the ions’ cyclotron motion is excited by radio frequency signals on electrodes, which are inserted through the observation slots of the inner drift tube. Similarly, the image charges of the revolving ions are picked up and Fourier-analyzed to determine the mass-over-charge ratios of the stored ions. These Fourier transform - ion cyclotron resonance (FT-ICR) measurements [3,4] have included the determination of the number of ions as well as the ion storage time. The coherence time of the ICR-induced collective ion motion was found to be of the order of 1 to 2 ms, allowing a charge-to-mass resolution of about \(5 \times 10^{-5}\). The validity of the method was checked by comparing the relative abundance of different charge states as found by FT-ICR and by x-ray techniques [8,9]. Similarly, the relative abundance of different isotopes was measured and compared to known ratios. In addition, the radial extent of the ion cloud was inferred.

Future progress will require to overcome a number of technical obstacles: For one, broadband excitation is not as easily accomplished as in conventional FT-ICR MS, since the range of cyclotron frequencies that have to be addressed is much wider for highly charged atomic ions. A typical FT spectrum obtained following broadband excitation is shown in Fig. 1. The signals from isotopically pure heliumlike through bare krypton ions are observed between 18 and 20 MHz. (Note that signals from light ions, such as bare nitrogen or oxygen, are missing, which illustrates the
displacement of low-Z ions by higher-Z ions in the ETM.) The figure illustrates another technical obstacle, i.e., a relatively large noise pickup in spite of the use of the shortest leads possible (by direct radial arrangement out of the trap). The noise level changes throughout the day and cannot easily be subtracted. Frequency shifts are expected due to relativistic effects at those higher cyclotron frequencies [10, 11]. These combine with the radial inhomogeneity of the magnetic field. The main obstacle, however, is the typical EBIT drift-tube construction where only small excitation and detection electrodes can be inserted (and only one for each function) in contrast to the much larger excitation and detection electrodes (i.e., differential electrode pairs) in use in conventional FT-ICR. We plan to redesign the trap’s middle drift tube into four segments. Each segment pair will be used for either excitation or detection. Note that similarly the Tokyo EBIT has recently been equipped with a middle drift tube with six segments.

![FT-ICR spectrum of krypton ions as produced in the electron mode by a 130-mA, 80-keV electron beam: (a) following broadband excitation by sweeping from 23 MHz down to 17 MHz within 100 μs. (b) noise spectrum in the absence of excitation.](image)

The FT-ICR studies laid the foundation for further investigations and uses of the magnetic mode. For example, why not have a look at the x-ray and optical signals, just as in the ETM? There are two sources of excited states available in the MTM as described in the following sections: (a) Excited metastable levels that have been populated in the electron mode just before the electron beam is switched off. (b) Electron transfer reactions from neutral atoms or molecules during the MTM.
ATOMIC LIFETIME MEASUREMENTS

Most atomic levels of highly charged ions have lifetimes in the picosecond range and are thus best studied by beam-foil spectroscopy. However, there is a class of levels that is far too long-lived for this traditional technique, but is of interest for the diagnostics of astrophysical and laboratory plasmas: the fine structure levels of the ground configurations of highly ionized atoms and similar levels in low-lying configurations that – for reasons of parity – cannot decay by the emission of electric dipole (E1) radiation. These levels are easily excited even in thermal plasmas, but their emission by higher-multipole order radiation can be observed only if they are not collisionally quenched, that is in environments of such low particle density as in the solar corona, tokamak discharges, or ion traps operating under ultra-high vacuum conditions.

The same electron beam of EBIT that produces highly charged ions is available for collisional excitation and de-excitation. When these processes suddenly end, one can follow the signal on a given spectral line with time and thus obtain the lifetime of the upper level. The Livermore EBIT group has pioneered such lifetime measurements, first by a modulation of the electron beam energy around the excitation threshold of the level of interest, e.g. the 1s2s \(^3S_1\) level in He-like Ne\(^{8+}\) ions, and recently by switching the beam off completely [12,13]. Some of the subsequent work on the same level in He-like ions of N through Mg, covering level lifetimes from some 30 \(\mu\)s to several milliseconds, provides the most stringent test (better than 0.5%) on the exceedingly accurate theoretical predictions.

![Image](attachment:image.png)

**FIGURE 2.** X-ray observation of the M1 decay of the lowest triplet level in He-like Ne ions [14]. Left: Excitation just above threshold, right: Excitation energy sufficient to also produce H-like and bare Ne ions. The curve tail results from CX (recombination).

In addition to these x-ray observations, a second realm of atomic lifetime determinations are the electric-dipole „forbidden“ M1 and E2 transitions that occur
in the visible spectrum and that are responsible for the long-time mysterious corona lines in the spectrum of the sun. Lifetimes in the range up to 15 ms have been measured for electronic states of Ar [15], K and Kr ions. Because of the poorer signal-to-noise ratio of detectors for visible light (in contrast to x-ray detectors), these measurements have reached a routine accuracy of „only“ 2% - which however is sufficient to exceed the intrinsic accuracy of the current theoretical predictions of in-shell M1/E2 transition rates by an order of magnitude.

Note that for any lifetime measurement a good characterization of the MTM is essential. In particular, the ion storage time and ion cloud dynamics need to be well known, lest the ions drift out of the solid angle subtended by the photon counters, reducing the apparent radiative lifetimes. For millisecond lifetimes, the correction for the finite ion confinement time has to be established, e.g., by employing charge exchange rate measurements, as is discussed in the next section.

ELECTRON TRANSFER REACTIONS

After the electron beam has been switched off, any delayed emission from the decay of short-lived levels signifies level population by charge exchange (CX) processes. With spectrally resolved photons, the individual ionic charge state reached after CX can be determined, and CX can be mapped for a number of charge states simultaneously. A typical spectrum illustrating the CX-induced K-shell emission in the magnetic trapping mode is shown in Fig. 3.

![FIGURE 3. Argon K-shell x-ray emission observed in ETM and MTM (from ref. 16).]
While CX measurements are interesting with respect to atomic [16] and space physics [17], and gas pulses may be applied to the trap volume for that purpose deliberately [8], they also provide information on the magnetic trapping mode. In recent EBIT-II experiments, rather similar effective confinement time constants (storage times of order 300 ms) were found by direct x-ray observation of He-like Kr ions and in the decay curve tails (cascades) of optical measurements on Kr ions with partly filled 3p and 3d shells, indicating that under the vacuum conditions in EBIT-II the confinement time in the MTM depends only weakly on the ion charge. For SuperEBIT, with its better vacuum, ion storage times of more than 20 s were measured. This showed that CX is the dominant loss process in the MTM. Other loss processes, such as radial diffusion or scattering into the axial loss cone, are much slower.

In addition, the ion storage time was found to depend on the trap potential that confines the ions in the axial direction. High potentials reduced the ion confinement time. We attribute this to the fact that the ions attain a higher temperature during the electron mode when confined in a deeper well [5], which in turn results in an increased likelihood for CX due to the higher ion velocity.

CONCLUSION AND OUTLOOK

The deployment of the MTM of EBIT has only just begun. Three major directions of study have been identified: Mass spectrometry, lifetime determinations and charge exchange measurements. Future extensions may include precision MS of known ions (as compared to mass analysis of unknown ensembles), coincidence measurements of decay cascades after CX, or the determination of the charge state of the previously neutral atoms after the reaction by MS, either by FT-ICR of the ions in the trap or after ejection and (time-of-flight or magnetic-sector) MS. So far the CX partners have been neutral atoms of noble gases. An obvious extension is the use of more and more complex reaction partners: small molecules, larger molecules, atomic and molecular clusters. Finally, it may be worth noting that by switching from the electron to the magnetic trapping mode, the highly charged ions are essentially “transferred” (in time) from their source to a Penning trap without the need of two (spatially) separated devices and a transfer line. Thus, in principle any EBIT/re-trap type of experiment may be reconsidered with the magnetic trapping mode as an alternative.
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