

Observation of Visible and uv Magnetic Dipole Transitions in Highly Charged Xenon and Barium

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We have observed an unusual transition which is predicted to result in visible and near-uv emission from very highly charged titaniumlike ions spanning the entire upper half of the periodic table. Measurements of the wavelengths of the $3d^4\ ^5D_{2-5}D_3$ transitions in Ba^{+34} and Xe^{+32} are in surprisingly poor agreement with *ab initio* calculations. This work was carried out in an electron beam ion trap and demonstrates that such a device can be an important tool for visible spectroscopy of highly charged ions.

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Although ions having identical numbers of electrons display similar spectral structure, the scale of many atomic properties changes dramatically as the nuclear charge (Z) is increased along an isoelectronic sequence. One example is the rapid decrease in transition wavelengths leading to the characteristic emission of light in the x-ray region of the spectrum for very highly charged ions. It was therefore unusual when Feldman, Indelicato, and Sugar [1] reported calculations predicting a small set of observable visible and near-uv magnetic dipole ($M1$) lines for highly charged ions in the titaniumlike isoelectronic sequence. The upper levels of these transitions lie within the ground term, and, therefore, in low density plasmas ($\leq 10^{13}\text{ cm}^{-3}$), where competition from electron collisions is negligible, a large fraction of excited levels should decay to them and result in intense forbidden lines. Even more intriguing is the fact that these lines are predicted to remain in the visible or near-uv region of the spectrum for a range of ions covering the entire upper half of the periodic table ($45 \leq Z \leq 92$). To our knowledge, this is the only candidate sequence of strong lines with visible or near-uv wavelengths persisting over the 1 to 10 keV range of ionization energies. Once measured, the lines are easily located even with low resolution spectrometers. In this Letter, we report the first observation and measurement of these unusual lines.

Interest in visible and near-uv forbidden transitions in highly charged ion is not only academic; there are important practical applications, particularly in high temperature plasma diagnostics. Long wavelength forbidden transitions have been seen in the solar corona [2,3] and in tokamaks [4-7], and they have been used extensively to probe a variety of properties for both kinds of plasmas. For example, local ion temperatures and bulk plasma velocities can be inferred from the substantial Doppler widths and shifts of these lines [5,6,8]. It is also possible to determine the direction and magnitude of internal magnetic fields from polarization measurements or from Zeeman shifting or broadening of the line profiles [9-11]. The

visible and near-uv transitions are preferred, since they can usually be measured more accurately and because refractive optics and polarizers can be efficiently employed. Most of the visible and near-uv diagnostic lines used to date come from ions with the ns^2np^k ($n = 2, 3$) ground configurations [12]. Unfortunately, these lines are expected to move to much shorter wavelengths in the high- Z (high ionization energy) elements of the sequence appropriate to the next generation of high temperature fusion devices. Thus, such lines will no longer be as easy to measure and will lose much of their value as diagnostic tools. The high radiation levels expected near these new devices will make remote diagnostic using fiber optics desirable, thereby increasing the need for visible or near-uv diagnostic lines.

In the present work, ions were created at the joint National Institute of Standards and Technology and Naval Research Laboratory Electron Beam Ion Trap (NIST-NRL EBIT) [13]. The trap is based upon the design of the Lawrence Livermore National Laboratory EBIT [14] and consists of three cylindrical drift tubes through which nearly monochromatic ($\sim 50\text{ eV}$ spread) and intense ($\sim 2000\text{ A cm}^{-2}$) electron beam passes. The electron beam is focused to a small diameter ($\sim 60\ \mu\text{m}$) by a 3 T axial magnetic field, and the electron energy is determined by the potential applied to the center drift tube. The ions are trapped axially along a 2 cm length by biasing the end drift tubes 250 V above the center drift tube; they are confined radially by the space charge potential of the electron beam and by the magnetic field. Injected atoms are converted to ions via collisions with the electron beam. A specific charge state can be preferentially produced by tuning the beam energy to just below its ionization potential. This capability for producing and trapping very highly charged ions in a small-scale laboratory device has made the EBIT a valuable source for x-ray spectroscopy [15]. The low effective electron density inside the EBIT ($\sim 10^{12}\text{ cm}^{-3}$), however, also makes it ideal for studying weak, forbidden transitions

within the ground terms of highly charged ions. We have now demonstrated this potential by using the EBIT for visible and near-uv spectroscopy of such transitions.

Barium and xenon were studied because they are easily introduced into the EBIT. Barium is emitted continuously from the heated cathode of the electron gun, and xenon can be loaded by directing a stream of neutral gas into the trap region through a side port. The electron beam energy required to maximize production of titaniumlike barium and xenon ions in the EBIT is 2.259 and 2.026 keV [16], respectively. Although the electron beam energy is determined by the center drift tube potential, the exact energy is somewhat less than this potential due to beam space charge defects. For a 35 mA beam, we estimate this space charge correction to vary from 72 to 63 eV as the center drift tube potential is varied from 1880 to 2570 V. For a higher current beam, the space charge correction is proportionately higher.

Light emitted from the trap was imaged by a pair of lenses onto a 250 μm entrance slit of a 0.25 m focal length, $f/3.5$ Ebert scanning monochromator. The magnification factor of the lens system was 0.8. A blue-sensitive photomultiplier was mounted behind a 500 μm exit slit and operated in photon-counting mode. With a 2360 grooves per mm grating, the instrumental width of the monochromator was 0.8 nm. The Doppler width of the spectra from the trapped ions is estimated to be less than 0.08 nm, and any Zeeman shifts less than 0.04 nm.

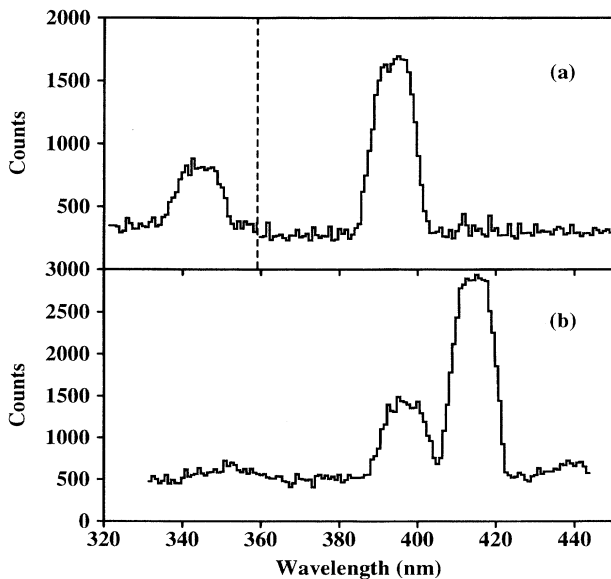


FIG. 1. Broad survey spectral scans with high (10 nm, monochromator slits removed) bandpass for (a) the $3d^4\ ^5D_2\text{-}^5D_3$ transition in Ba^{+34} (right peak) and the $3d^5\ ^4G_{7/2}\text{-}^4G_{9/2}$ transition in Ba^{+33} (left peak), and (b) the corresponding transitions in Xe^{+32} and Xe^{+31} . The EBIT drift tubes were set at 2378 and 2277 V for the portions of the spectrum in (a) to the right and left of the dotted line, respectively, and at 2144 V for the entire spectrum in (b). The electron beam current was 50 mA throughout.

The wavelength range studied here (320 to 443 nm) was limited at the lower end by the diminishing transmission of the lenses. Calibration of the monochromator's wavelength scale was performed by shining light from a number of low pressure discharge lamps, including mercury, helium, neon, argon, and xenon, through the EBIT from a port opposite the spectrometer. Argon and krypton were also injected, ionized, and trapped during the course of the experiment to observe previously reported [12,17] $M1$ lines *in situ*. In this case, a relatively dense stream of gas was flowed continuously into the EBIT. This broadened the charge state distribution by providing a significant flux of neutral atoms from which steady state populations of the lower charge states, specifically Ar^{+13} and Kr^{+22} , could be formed. Such low charge state ions could not otherwise be made, since the EBIT electron beam became unstable at energies below 1 keV. Our *in situ* measurement of the krypton line yielded 384.08(20) nm, and that of the argon line yielded 441.32(20) nm. These are to be compared with their literature values [12,17] of 384.09(3) nm and 441.24(2) nm, respectively. We estimate the overall uncertainty in our wavelength calibration to be ± 0.2 nm.

Initially, broad survey scans (340–440 nm) with wide spectral bandpass (10 nm, monochromator slits removed) were conducted (Fig. 1). These were followed by narrower scans in the regions where prominent spectral features appeared (e.g., Fig. 2). Finally, the monochromator was adjusted to the peak of the observed lines, and the magnitude of their signal was recorded as a function of electron beam energy. The well controlled conditions in the EBIT produced only a few lines in the survey spectra and greatly simplified the task of identifying them.

There are no previously measured and identified lines from titaniumlike, vanadiumlike, and neighboring $3d^N$ shell ions of barium and xenon. It was not possible, therefore, to confirm the presence of these ions in the trap by observing an experimentally known transition. The existence of trapped barium and xenon was confirmed,

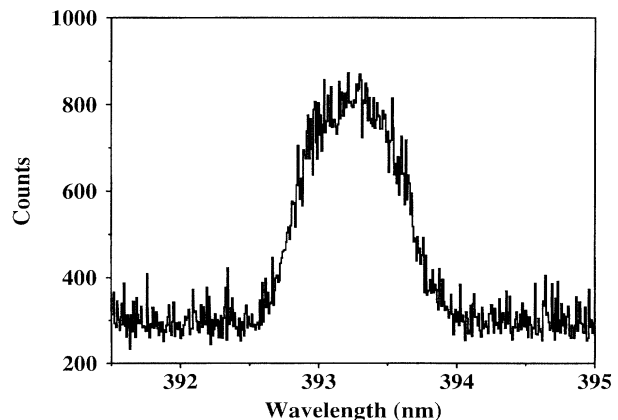


FIG. 2. $3d^4\ ^5D_2\text{-}^5D_3$ transition in titaniumlike Ba^{+34} . The profile shown is the sum of 10 individual scans, each of which was accumulated in 5 min with a drift tube potential of 2378 V.

however, by the observation of known x-ray lines from a higher charge state (Ne-like) in high-resolution spectra [18] at increased electron beam energies. Furthermore, the features seen in low-resolution x-ray spectra, taken simultaneously with our uv data, were consistent with theoretical predictions [we used the GRASP² multiconfiguration Dirac-Fock (MCDF) code] [19] for radiative recombination into, and transitions between, levels in $3d^N$ shell ions of Ba and Xe. These were the only charge states of Ba and Xe whose predicted features were consistent with the observed low-resolution spectra.

In Fig. 3, we show energy level diagrams for the barium ions produced in our study. The corresponding diagram for xenon ions, although not shown, is very similar. After a tentative identification of the prominent spectral feature at 393.24 nm (Figs. 1 and 2) as the titaniumlike barium line, whose wavelength was originally predicted [1] to be 377.8 nm, we used the Cowan relativistic Hartree-Fock (HFR) atomic structure code with electron correlation correction [20] to improve the prediction. For the $3d^N$ configuration, the two Slater integrals, $F^2(3d3d)$ and $F^4(3d3d)$, and the $5d$ spin-orbit integral (ζ) determine the energy level positions. We scaled the F^k parameters to make the code's predicted wavelength match that of the tentatively identified line exactly. This scaling factor turned out to be 93% of the *ab initio* HFR values. This is a relatively modest adjustment since scale factors between 90% and 95% are typically required for highly charged ions [20]. Using the 93% scaling, a calculation of the $3d^4 5D_2-5D_3$ transition in titaniumlike Xe⁺³² gives a value of 409.4 nm. This is within 5 nm of a strong line recorded during Xe injection, with the electron beam energy tuned to produce Xe⁺³². As presented in Fig. 3, the $J = 3$ level can decay via the $J = 2$ or the $J = 4$ levels. Calculations using the Cowan code indicate that the A values for the $J = 2-3$ transition in xenon and barium are approximately 400 s^{-1} . The A value for the $J = 3-4$ transition, however, is only 2 s^{-1} in xenon and 17 s^{-1} in barium. Therefore, most of

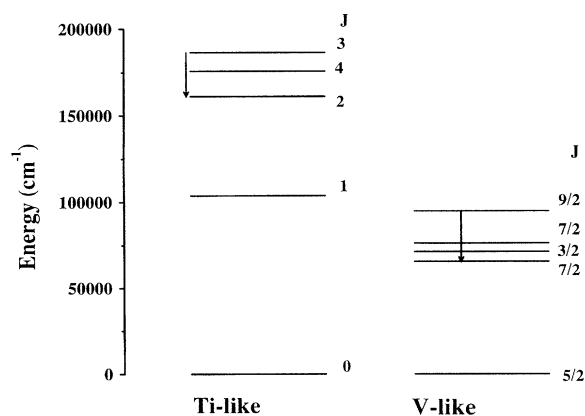


FIG. 3. Energy level diagrams of the lowest levels in the ground terms of titaniumlike and vanadiumlike barium. The level energies were calculated using the Cowan code and arrows indicate visible and near-uv $M1$ transitions.

the decays from the $J = 3$ level will proceed via the $J = 2-3$ channel, and the transition should be easily observable under our plasma conditions.

As a separate check on the scaling factor, we calculated the $3d^8$ levels of ironlike Sn⁺²⁴, the closest highly charged ion of the $3d^N$ shell ($2 \leq N \leq 8$) in which the energies of all the levels of the ground configuration are experimentally known. Using the same 93% F^k parameter scaling for the $3d^8$ configuration, we find agreement with the previously measured values [21] to within 1%.

After observing the titaniumlike barium line, we conducted a theoretical search for other observable $M1$ transitions arising from the $3d^N$ ground configurations. This involved completing calculations for the other charge states of barium and xenon from calciumlike to ironlike ions, using the 93% scaling of the F^k parameters. By examining the calculated wavelengths and branching ratios, we predicted that only one more line should be observable, specifically a $3d^5 4G_{7/2}-4G_{9/2}$ transition in vanadiumlike barium and xenon. The wavelength of this line is predicted to decrease rapidly with increasing Z and to leave the near-uv spectral range ($\geq 300 \text{ nm}$) for ions with $Z \geq 58$.

These predictions prompted us to take a survey scan at shorter wavelengths, with the electron beam energy tuned to produce vanadiumlike barium. A single prominent feature was found, and the wavelength fell very close to the calculation (Table I). Later, when a survey scan was made in xenon, the vanadiumlike line was also found near the predicted location. Thus the wavelengths of all four prominent features recorded in the survey spectra matched the scaled theoretical values closely (Table I).

It should be noted that the noble atomic gases (argon, krypton, xenon) were introduced separately into the EBIT, with sufficient time between loadings to allow the previous gas to be removed by a combination of cryogenic and ion pumping. Features associated with ions of any one of these species were not seen in the spectra of the others. Further evidence for the proper identification of these lines came from the measurement of their ampli-

TABLE I. Predicted and measured wavelengths of visible and near-uv $M1$ forbidden transitions in titaniumlike Ba⁺³⁴ and Xe⁺³² and vanadiumlike Ba⁺³³ and Xe⁺³¹.

Ion	Transition	Wavelength ^a (nm)		A^c (s^{-1})
		Calculated ^c	Measured	
Ba ⁺³⁴	$3d^4 5D_2-5D_3$	393.2 ^b	393.24 ± 0.20	433
Xe ⁺³²	$3d^4 5D_2-5D_3$	409.4	413.94 ± 0.20	434
Ba ⁺³³	$3d^5 4G_{7/2}-4G_{9/2}$	341.4	343.57 ± 0.20	425
Xe ⁺³¹	$3d^5 4G_{7/2}-4G_{9/2}$	394.3	396.25 ± 0.20	311

^aWavelengths in air.

^bThe F^k scaling in the Cowan code was adjusted to 93% so that the calculated and measured wavelengths of this line were equal.

^cCalculated with Cowan code using a 93% scaling of the $F^2(3d3d)$ and $F^4(3d3d)$ Slater integrals.

tudes as a function of electron beam energy. Because the ionization potentials of the nearby charge states are only separated by about 100 eV in both barium and xenon, the populations of these various charge states, and hence the signal strengths of the corresponding emission lines, are expected to change quickly with beam energy. This was seen in our measurements (e.g., Fig. 4) where it was found that all of the identified lines exhibited the expected behavior with electron beam energy, following the predicted [22] population densities of their parent ions.

Although our scaled HFR calculations reproduce the experimental data quite well, a state-of-the-art, fully *ab initio*, MCDHF calculation [23] fails to yield good agreement [16]. To our knowledge, there is no other case in which a fully relativistic calculation disagrees with transition energies of highly charged ions ($q \geq +30$) by as much as 5%. The measurements reported here therefore present an important challenge to theorists seeking to predict accurately atomic structure from first principles.

In summary, we have observed a special set of lines from highly charged ions which are expected to persist in the visible and the near-uv range of the spectrum over a large range of nuclear charge states. It is anticipated that this will have important practical application for remote diagnosis in the next generation of high temperature plasma fusion devices. Measurement of these lines also provides a sensitive probe of atomic structure theory applied to a range of ions where the accuracy has been limited by a scarcity of experimental data.

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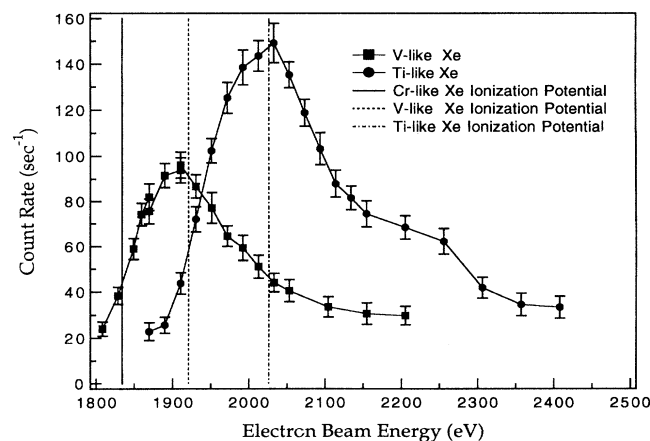


FIG. 4. Amplitudes of the $3d^4 5D_{2-5}D_3$ observed titaniumlike and $3d^5 4G_{7/2-4}G_{9/2}$ vanadiumlike transitions in xenon versus electron beam energy, for 35 mA of electron beam current. The corresponding ionization energies are shown by vertical lines.

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