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# FORBIDDEN LINES FROM HIGHLY CHARGED, METASTABLE ION BEAMS

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# Forbidden Lines from Highly Charged, Metastable Ion Beams

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Forbidden lines have been observed in emission from intense, continuous beams of metastable, highly charged ions produced by an electron cyclotron resonance ion source. The lines are magnetic dipole transitions between levels in the ground configurations of ions with incomplete 2p or 3d subshells. Wavelength measurements have been made for lines from the $2p^5$ ions $Ar^{9+}$, $K^{10+}$, and $Ca^{11+}$, the $2p^4$ ions $K^{11+}$ and $Ca^{12+}$, and the 2p ions $Ar^{13+}$ and $K^{14+}$. Measurements have also been made of lines from transitions between levels of the $3d^{9,8,7}$ ions $Nb^{14,15,16+}$. All of the lines lie in the visible spectrum. The technique makes use of a position sensitive photomultiplier at the exit of a 0.25m spectrometer.

Observations of forbidden emission lines can provide important diagnostic information on astrophysical and fusion plasmas (e.g. in the solar corona, flares and tokamak devices). This results from their sensitivity to electron density and temperature and from the fact that these lines often lie in the visible or near ultraviolet spectrum where efficient transmitting and reflecting optics can be used.

Measurement of forbidden line wavelengths has intrinsic value in establishing structure in the lowest lying electron configurations. For most highly ionized systems, this structure is known only from observations of allowed transitions which, because of their large wavenumbers, provide low precision when the differences are taken to derive the spacing of the low lying metastable levels. Thus a direct measurement of a forbidden line with moderate precision can exceed in accuracy a value determined from allowed lines which were measured with much higher precision. Conversely, direct determination of forbidden line wavelengths can establish, with high precision, energy intervals (or scale) in the XUV or X-ray region where the allowed lines, which connect to the metastable levels, fall.

Earth bound sources of forbidden emissions from highly charged ions have been largely limited to the fusion plasmas and foil excited beams of fast heavy ions. The latter have been especially valuable in study of the forbidden decay modes of H-like and He-like ions. This work reports observations of several forbidden lines from a new source of such emissions, namely low velocity multiply charged ion beams from the Lawrence Berkeley Laboratory (LBL) Electron Cyclotron Resonance (ECR) ion source[1,2]. The ECR ion source and beam transport line are located on the vault roof of the LBL 88-inch cyclotron. The source provides heavy ions for injection into the cyclotron, and is also used for atomic physics research with the slow beam before acceleration. The ECR source creates the excited ion beams studied in this work (no exciting medium such as a gas cell or thin foil is used) and they are charge analyzed before observation; thus there is little ambiguity as to which ion is being observed.

## I. Method

The results reported here were obtained by viewing a portion of the ion beam at a position about 5 meters from the ion source. Radiation emitted by the decay of metastable ions in the beam is analyzed with a small (0.25 meter) spectrometer which is equipped with a position sensitive photomultiplier mounted at the exit plane. Figure 1 is a sketch of the arrangement. The spectrometer is an f/3.6 instrument and is oriented so that the central ray through the entrance slit is normal to the ion beam. The Doppler broadening of emission lines from the beam ions is then in the range of 0.2 to 0.4 nm full width (all beams had energies of 10KV times the ion charge). With the entrance slit at 0.5 mm, the instrument width is 1.7 nm and 0.8 nm using the 1180 groove/mm (above 450 nm) and the 2360 groove/mm (below 450 nm) gratings, respectively. Wavelength calibrations were made for each line observed by recording spectra from hollow cathode lamps containing Ba, Fe, Mo, and Ne carrier gas immediately preceding or following collection of the unknown lines, and without readjustment of the spectrometer. The hollow cathode lamps were mounted opposite the spectrometer as shown in Figure 1.

The position sensitive photomultiplier and associated electronics provides two output pulses for each photo event. The amplitudes of these pulses are a measure of the $x$ and $y$ position from which the photoelectron originated at the photocathode. The detector was oriented with $y$ axis parallel to the spectrometer dispersion, and the $y$ pulses were histogrammed with a standard multichannel pulse-height analyzer. This method of data collection is simple to implement, but causes some asymmetry in a spectral line because of the curvature of the slit image in the exit plane. (The same distortion is present with a scanning instrument and a straight exit slit.) This effect leads to lines which are broad on the long wavelength side and is present with the unknown and calibration lines. The asymmetry does not contribute a significant systematic shift in line center determinations at the level quoted here. A more satisfactory scheme would remove the image curvature via computer processing of the $x,y$ data. This would narrow the lines somewhat and improve the peak signal-to-noise ratio.

At a distance $z$ from the ion source, the beam particle current, $I_i$, of metastable ions in state, $i$, is given by

$$I_i = w_i I_o e^{-z/v\tau_i} \tag{1)}$$

where $w_i$ is the fraction of the total beam particle current, $I_o$, which is in metastable state $i$. The lifetime of the metastable state is $\tau_i$, and $v$ is the beam velocity. A given transition $j-i$ with radiative decay rate $A_{ji}$, will yield a photon emission rate per unit length of beam path given by

$$R_{ji} = A_{ji} I_i / v. \tag{2)}$$

At 10 KV extraction potential, the beam velocities from the ECR source are in the range of 0.5 to 1.0 x $10^8$ cm/sec, so that the transit times to the spectrometer are in the range of 5-10 $\mu$sec. This is much longer than the lifetimes of excited states which decay by allowed electric dipole radiation ($\leq 10^{-8}$ sec), but far shorter than typical metastable lifetimes ($\tau_i \approx$1–100 msec). Thus at the location of the spectrometer, the beam should be dark except for the forbidden emission lines.

Since the decay length ($v\,\tau_i$) of the metastable ions is in the range 0.5 to 100 km, and the spectrometer is at $z \approx$5m, the exponential in Eq 1) is essentially unity. The spectrometer views about 10 cm of beam path so only a fraction $10^{-6}$ to $10^{-4}$ of the metastable ions decay in this region. This tiny fraction is compensated for by the intensity of the ion beams and the efficiency of position sensitive photon counting at the spectrometer exit.

Taking the magnetic dipole (M1) transition in F-like (9 electron) $Ar^{9+}$, $2p^5\ {}^2P_{3/2-1/2}$, as an example, with $A_{ji}$ =130 $sec^{-1}$ and a beam current (electrical) of 80 $\mu$A, $I_o$ = 5 x $10^{13}$ $sec^{-1}$, $v$ = 8 x $10^8$ cm/sec and, using the statistical value $w_i$ =1/3, one has $R_{ji} \approx$ 2 x $10^6$ $sec^{-1}cm^{-1}$. Thus over a 10 cm beam length, one has about 2 x $10^7$ photons/sec available into $4\pi$ solid angle. When one includes the spectrometer thoughput and detector efficiency (that is, one does the experiment) a net signal count rate of about 0.1 $sec^{-1}$ is observed. This rate provides good signal-to-noise with integration periods of 15 to 30 minutes. Although the signal rate is low, so is the effective dark rate of the photomultiplier since the total dark "flux" of some 10 counts/sec is spread uniformly over the 25 mm diameter photocathode, whereas the forbidden line image is concentrated on a small area. The effective dark rate under the line is typically about 0.02 counts/sec. This line is shown in figure 2. The futility of attempting these observations with a scanning spectrometer and an ordinary photomultiplier may be apparent.

The ion beam is charge analyzed before reaching the region viewed by the spectrometer, and thus, nominally one expects only forbidden lines from the selected ion to be present in the emission spectrum. This is often the case, however, (1) excitation of the residual gas molecules in the transport line by the beam ions, or (2) electron capture by the beam ions from the residual gas, can produce weak emission lines which can confuse the interpretation of a spectrum. Lines from the residual gas are largely insensitive to the incident ion charge, and lines from electron capture into highly excited states of the projectile are seen for charges differing by one unit from the selected charge state. Thus to assure identificatiion of a forbidden line with the selected ion of charge q, observations are made of the same spectral region with ions of adjacent charges (q±1). Common lines seen in these spectra are assumed to arise from processes (1) or (2); a line unique to the charge q is assigned to the forbidden spectrum of that ion. The residual gas pressure in the transport line is typically about 2 x $10^{-8}$ Torr, and is not easily varied in a controlled way.

## II. Results and Discussion

Using the technique outlined above, observations have been made of forbidden lines from F-like (9 electron) $Ar^{9+}$, $K^{10+}$, and $Ca^{11+}$; O-like (8-electron) $K^{11+}$ and $Ca^{12+}$, and B-like (5-electron) $Ar^{13+}$ and $K^{14+}$. Examples of these observations are shown in figures 2 and 3, and the results are summarized in Table I. The uncertainties quoted are about ± 1/10 of the line widths in most cases; lines observed with low signal-to-noise ratio (e.g. $K^{14+}$) have uncertainties of about ± 1/5 the line widths. The line centers were determined by a centroid routine applied to both the unknown and calibration spectra. The wavelength scale was then determined from a least squares fit of calibration line centers (expressed as data channel numbers) to a linear function of their wavelengths.

Many lines in these sequences have been observed from the sun or other hot astrophysical sources (Z less than about 26) and from tokamak plasmas (Z up to about 32). However, the results reported here are the first direct observations of these lines in the ions $K^{10,11,14+}$, and are the first laboratory observations of nearly all of the others listed in Table I. Because the number of observations of the $2p^n$ sequence lines is fairly large and extends over a wide range of Z values, quite accurate iso-electronic fits have been made by Edlén[3], Curtis[4] and others, which allow confident predictions of values for lines not yet observed in the range Z less than about 40. The recent compilation by Kaufman and Sugar[5] contains a summary of observations and predicted values. Ab initio calculations have been made by a number of workers and some of these are listed in Table I. The reader may note the excellent agreement of many of these calculations with the observations, however, this is somewhat misleading since it happens that the deviation of the computed values from measurements, if plotted versus Z, goes through zero near Z=18 (Ar). (See for example Refs. 10 and 11).

Recently, observations have been made of forbidden lines from some of the levels of the $3d^n$ ground configurations of $Nb^{q+}$ ions with $q=14,15,16$ ($3d^{9,8,7}$ Co, Fe and Mn sequences). Examples are shown in figure 4 and the wavelengths are listed in Table II. The observational base is far less extensive in the $3d^n$ sequences than in $2p^n$; there have been some direct observations[21] in Z=40 (Zr) and Z=42 (Mo) from the PLT tokamak, but the bulk of data on the ground configuration levels is from observation of allowed lines. Edlén[20] has made a sequence fit for the $3d^9$ $^2D_{5/2-3/2}$ and $3d^8$ $^3F_{4-3}$ lines, and his predictions for $Nb^{14,15+}$ are listed in Table II. The observation of the $3d^8$ $^3F_2$-$^3P_2$ transition is in agreement with the value obtained by Reader and Ryabtsev[18] from observations of $3p^6 3d^8$-$3p^5 3d^9$ lines from a low-inductance vacuum spark. The LS notation is used for these levels, but Reader and Ryabtsev's analysis indicates that $^3P_2$ contains about 30% $^3F_2$, which permits a magnetic dipole (forbidden otherwise) as well as electric quadrupole transition matrix element. The line in $Nb^{16+}$, $3d^7\ ^4F_{9/2-7/2}$, is in agreement with the prediction of Wyart[19] et al. which is based upon parametric fits to energy levels obtained from observations of allowed transitions.

The approach described here has potential for extending forbidden line measurements to a wide range of elements with charges extending up to the low 20's or so (with the present ECR source). The method relies upon the LBL ECR source's ability to produce and excite intense beams and the efficiency of position sensitive photon counting. Increased signal rates can be achieved by reducing the beam velocity which in turn may provide improved precision by allowing use of a higher resolution spectrometer. Extension into the vacuum ultraviolet region down to about 130 nm is feasible with the present detector.

## III. Acknowledgment

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Table I. $2p^n$ Forbidden Line Wavelengths (Å, air).

| Transition | Ion | Experiment[*] | Sequence Fits | Theory |
|---|---|---|---|---|
| $2p^5\ {}^2P_{3/2-1/2}$ | $Ar^{9+}$ | 5533.4(13)<br>5536.(40)[a]<br>5533.4(4)[b] | 5534.0[f]<br>5532.8[e] | 5534.6[g,j] |
| | $K^{10+}$ | 4255.1(9)<br>4260.1(36)[c,d] | 4256.8[f]<br>4255.6[e] | 4256.3[g,j] |
| | $Ca^{11+}$ | 3325.9(18)<br>3327.5(4)[k]<br>3327.9(22)[n] | 3327.1[f] | 3326.3[g,j] |
| $2p^4\ {}^3P_{2-1}$ | $K^{11+}$ | 5277.2(18)<br>5276.4(97)[f]<br>5274.5(55)[n] | 5278.6[f] | |
| | $Ca^{12+}$ | 4085.5(9)<br>4087.1(3)[f]<br>4092.7(33)[n] | 4087.0[f] | |
| $2p\ {}^2P_{1/2-3/2}$ | $Ar^{13+}$ | 4412.3(9)<br>4412.4(10)[b] | 4412.8[f]<br>4411.4[e] | 4411.8[h]<br>4413.8[i]<br>4421.2[j] |
| | $K^{14+}$ | 3445.3(13)<br>3446.5(32)[b,m] | 3446.3[f] | 3442.1[h] |

[*] The first entry for each transition is from this work.

[a] Ref 6, [b] Ref 7, [c] Ref 8, [d] Ref 9, [e] Ref 4, [f] Ref 3, [g] Ref 10, [h] Ref 11, [i] Ref 12, [j] Ref 13, [k] Ref 14, [m] Ref 15, [n] Ref 16.

Table II. $3d^n$ Forbidden Line Wavelengths (Å, air).

| Transition | Ion | Experiment[*] | Predicted |
|---|---|---|---|
| $3d^9\ ^2D_{5/2-3/2}$ | $Nb^{14+}$ | 4279.1(9)<br>4275.9(140)[a] | 4279.4[d] |
| $3d^8\ ^3F_{4-3}$ | $Nb^{15+}$ | 4772.0(9)<br>4769.6(110)[b] | 4769.0[d] |
| $3d^8\ ^3F_2-^3P_2$ | | 4552.3(18)<br>4554.5(140)[b] | |
| $3d^7\ ^4F_{9/2-7/2}$ | $Nb^{16+}$ | 5330.9(18) | 5335.(20)[c] |

[*] The first entry for each transition is from this work.

[a] Ref 17, [b] Ref 18, [c] Ref 19, [d] Ref 20.

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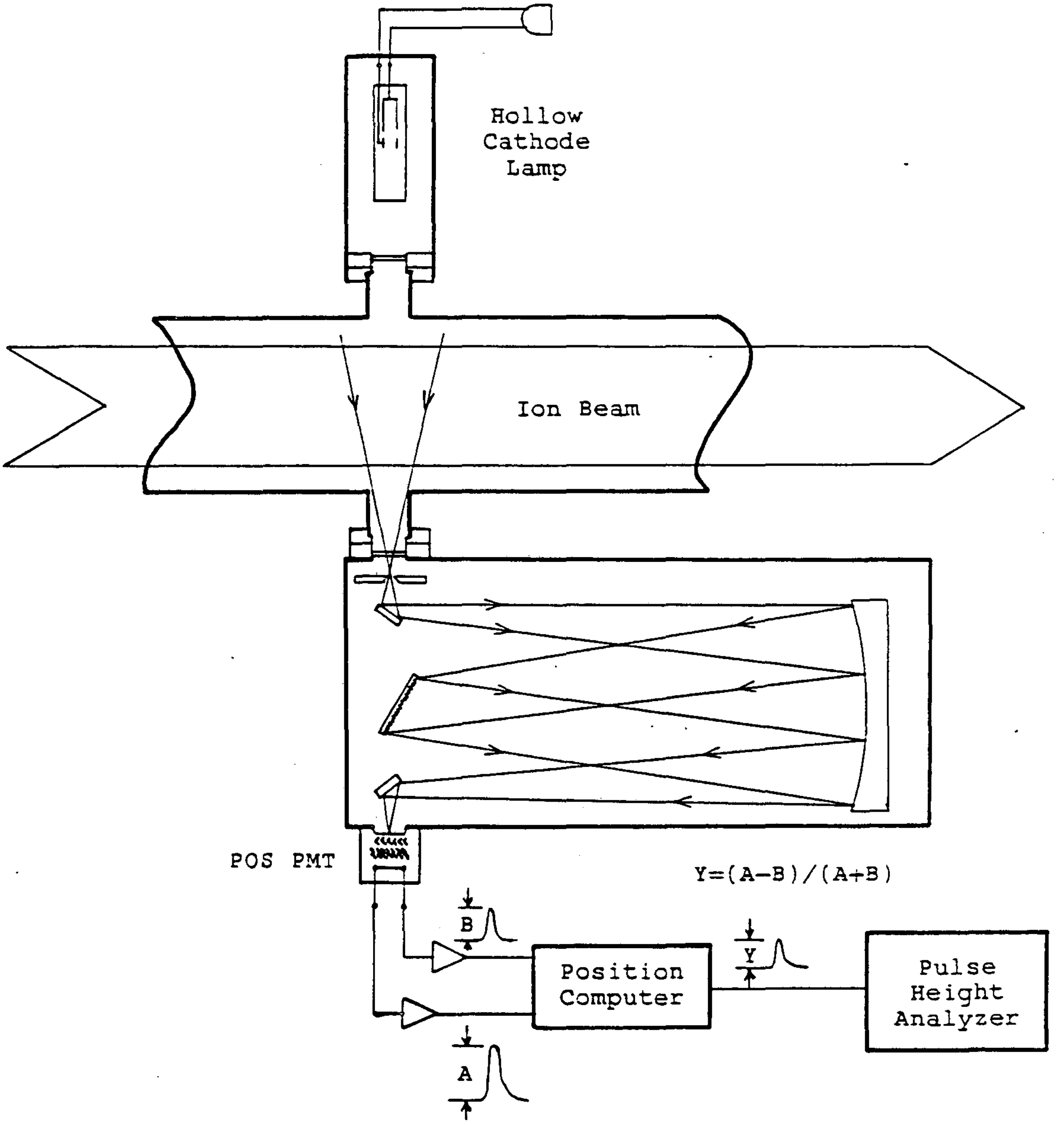

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Figure 1. Sketch of the experimental arrangement. The position sensitive photomultiplier (POS PMT) is adjusted so that its photocathode lies in the exit focal plane of the spectrometer with *y* axis parallel to the dispersion. The spectrometer is located about 5m from the ion source.

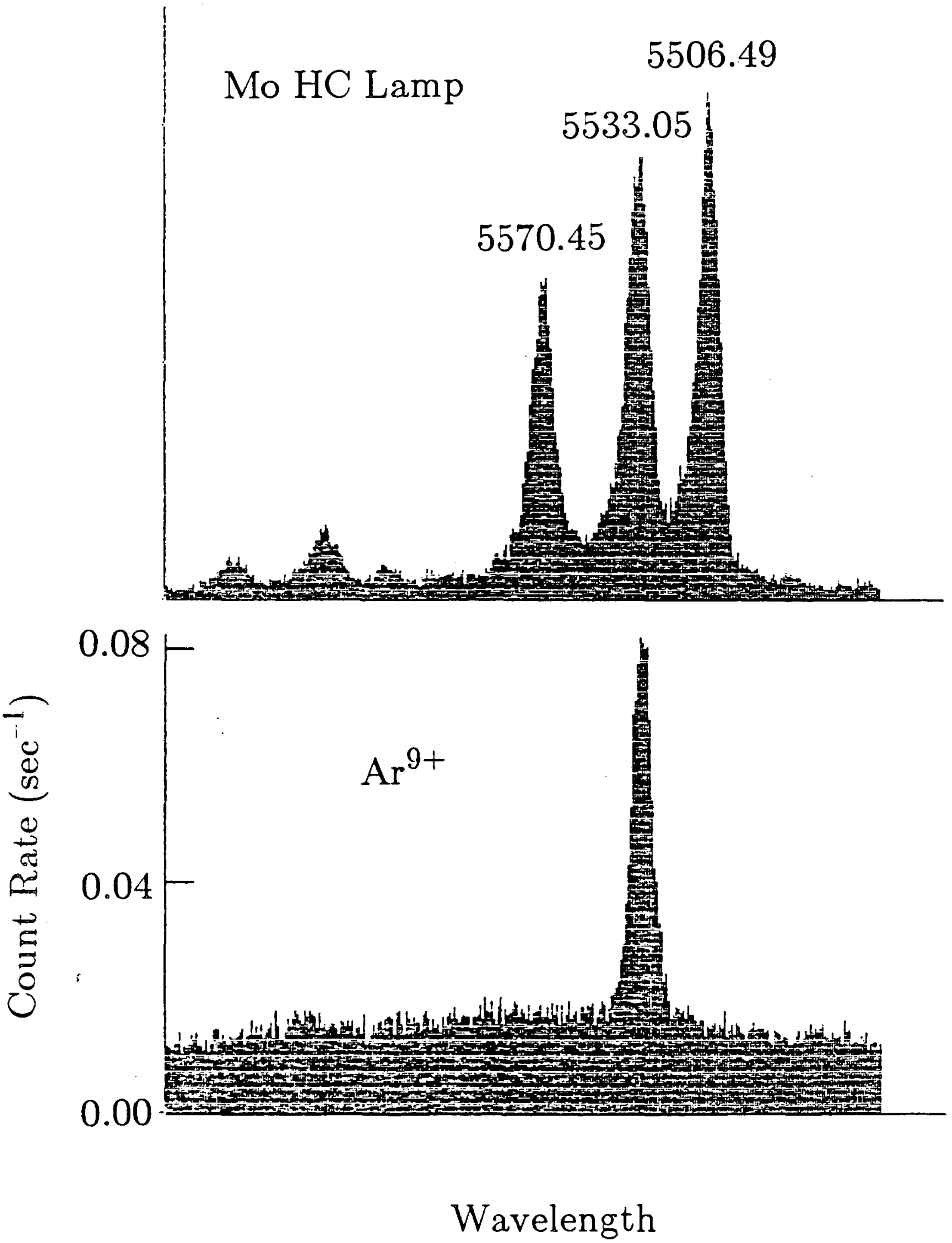

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Figure 2. The fine structure transition $2p^5\ ^2P_{3/2-1/2}$ in $Ar^{9+}$ (lower) and calibration lines from a Mo hollow cathode lamp (upper). The wavelengths are air values in Å. The $Ar^{9+}$ data were accumulated in about 30 min. from a beam of about 45 μA (electrical).

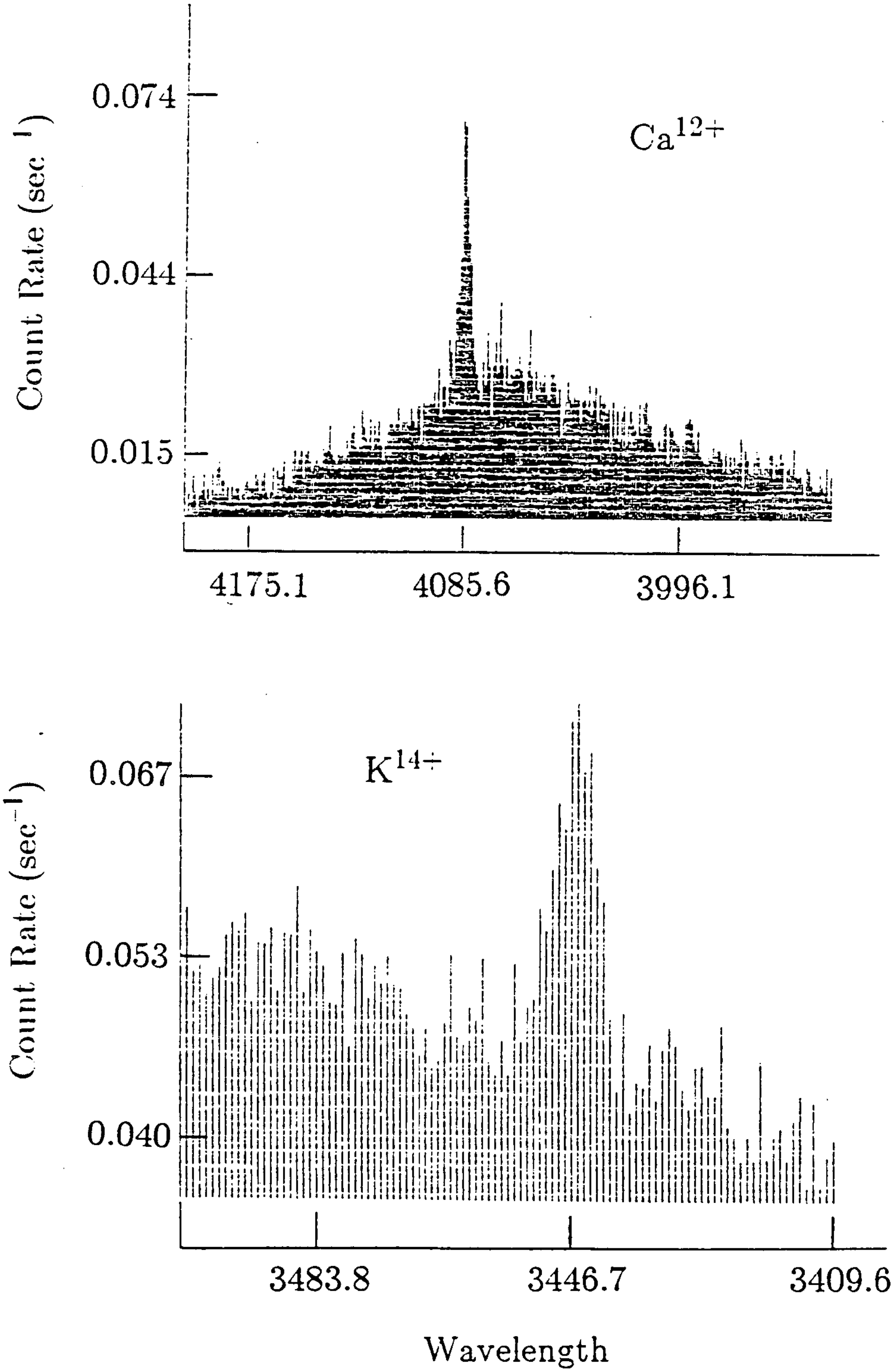

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Figure 3. The transitions $2p^4\ ^3P_{2-1}$ in $Ca^{12+}$ and $2p\ ^2P_{1/2-3/2}$ in $K^{14+}$.

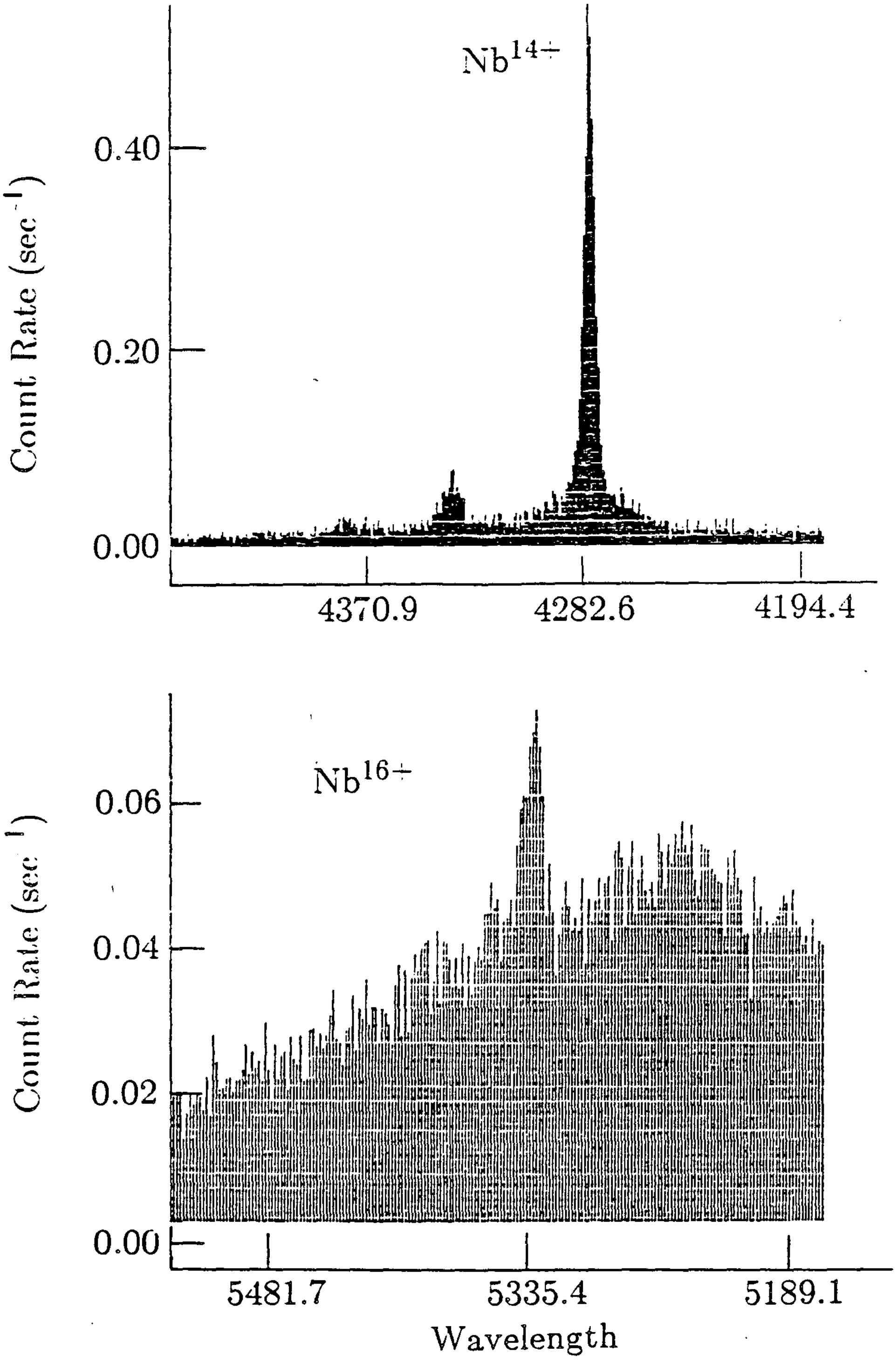

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Figure 4. The transitions 3p$^9$ $^2D_{5/2-3/2}$ in Nb$^{14+}$ (large peak) and 3d$^7$ $^4F_{9/2-7/2}$ in Nb$^{16+}$. The small peak to the left of the Nb$^{14+}$ line is from decay of excited states of beam ions which have captured an electron from the residual gas in front of the spectrometer.

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