

Using 'resonant' charge exchange to detect traces of noble gas atoms

J. E. Hardis, W. R. Peifer†, C. L. Cromer, A. L. Migdall, and A. C. Parr

Radiometric Physics Division, NBS, Gaithersburg, MD 20899

ABSTRACT: We describe an experiment in progress to measure the charge-exchange cross sections of Kr^+ incident upon Rb. We will measure the column density in the Rb cell using an optical interferometer. We expect this reaction to generate a significant flux of Kr atoms in the $5s [3/2]^o (J=2)$ metastable state, which will be useful as a step in RIMS studies of Kr isotope distributions.

1. INTRODUCTION

The advantages of resonance ionization in mass spectrometry (RIMS) are well known, but the technique is difficult for noble gas atoms because of their high ionization energy. In the present experiment, we seek to prepare noble gas atoms, such as krypton, in a metastable state (i.e., $5s [3/2]^o (J=2)$) so that resonant ionization may be achieved by using CW lasers. Our interest lies in methods that can be operated continuously (without the duty cycle losses inherent in pulsed laser schemes) and "on-line", that is, together with traditional methods of mass analysis. In this manner, we hope to reduce isobaric interferences while minimizing "off-line" enrichment as a separate step.

The process we are studying is illustrated in Fig. 1. It may be considered as a stage of filtering in a larger analytical instrument. Ions entering from the left are neutralized by charge-exchange collisions, forming metastable atoms. Any part of the beam not neutralized is swept away by electric field plates. Finally, the metastable atoms in the beam are reionized before they continue on to other elements in the instrument.

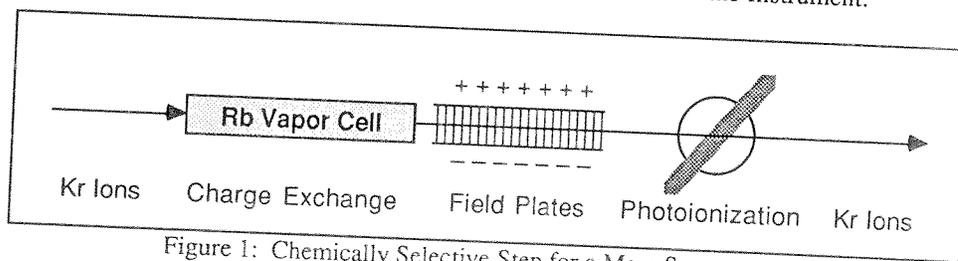


Figure 1: Chemically Selective Step for a Mass Spectrometer

Depending on the species of interest, this arrangement provides up to five different discriminations: resonant photoionization (chemical selectivity), Doppler shift tuning (velocity selectivity), hyperfine structure and isotope shift (isotopic selectivity), and the so-called "resonant" charge exchange, which also provides chemical selectivity.

2. "RESONANT" CHARGE-EXCHANGE

In Fig. 2, note that the binding energy of the valence electron of Rubidium almost equals the binding energy of the 5s electron in metastable krypton. (The J=2 metastable state has the lowest energy of the manifold and is the one labeled.) Because of this, the charge exchange scattering of Kr ions on Rb into Kr⁺ has been called "resonant", even though it isn't a resonance in the traditional senses.

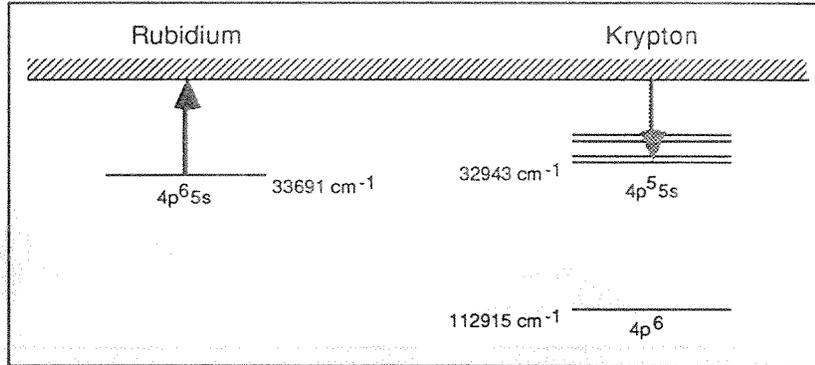


Figure 2: Energy levels of the Kr⁺ on Rb Resonant Charge exchange

This sort of process is well known for generating the metastable triplet states in He, and it has been used to a lesser extent to generate metastable states in other noble gases, such as Ne (Snyder and Hall 1975). However, to our knowledge, no experimental data exists for heavier noble gases such as Kr and Xe. Ice and Olsen (1975) predict large charge exchange cross sections, in excess of 70Å², but they don't report final state distributions. We expect to find large numbers of metastable Kr atoms formed by this process, perhaps as high as 20-40% of the total. A naive multiplicity argument suggests that about 5/12 of the beam that forms a 5s state directly will be in a J=2 state. This number will be reduced by other effects, such as Penning ionization caused by subsequent collisions. These large cross sections imply interactions at large impact parameters, which indicates that momentum transfer of the beam (angular deflection) should be a small effect.

3. CURRENT RESEARCH

Our current research is aimed at measuring the charge-exchange cross section of Kr ions on Rb. That is, we wish to measure the loss of ion current proportional to the vapor pressure in a charge-exchange cell (at low pressures). In Fig. 3, we outline the main elements in the experiment.

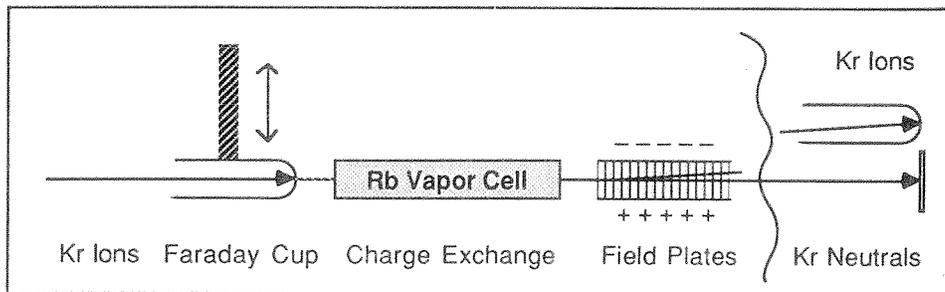
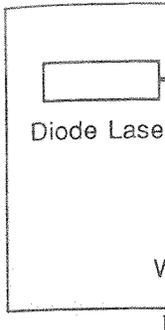


Figure 3: Schematic of the Charge-Exchange Experiment

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The other two plates in Fig. 4 811.5 nm laser

One of the difficulties in reporting an absolute cross section is knowing accurately the integrated *column density* in the vapor cell. The vapor pressure is higher in the heated center than at the ends, where the cell is cooled. However, we can infer the column density by measuring the optical path length of the cell near a Rb resonance line. The method is similar to the hook method of determining the oscillator strength of a transition.

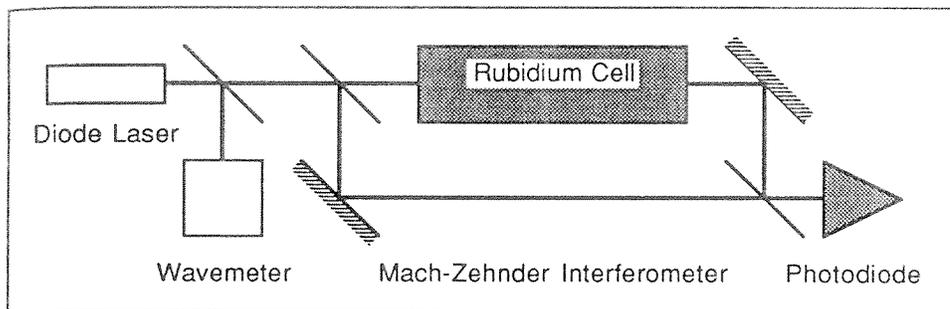


Figure 4: Measuring Rb Column Density Interferometrically

In Fig. 4, we show a Mach-Zehnder interferometer constructed around the charge exchange cell. We scan a semiconductor laser near the resonance line and look for interference beats on a silicon photodiode. The total light intensity on the photodiode can be represented as

$$I(\lambda)_{Total} = I(\lambda)_A + I(\lambda)_B + 2\sqrt{I(\lambda)_A I(\lambda)_B} \cos(\theta(\lambda))$$

where *A* and *B* refer to the two arms of the interferometer. In the interference term,

$$\theta(\lambda) = \frac{2\pi}{\lambda} \left(\frac{r_0 N f l \lambda_0^3}{4\pi(\lambda - \lambda_0)} \pm \epsilon \right)$$

where λ is the laser wavelength, λ_0 is the wavelength of the atomic resonance, r_0 is the classical electron radius (2.818×10^{-6} nm), the Nl product is the column density (vapor density times column length), f is the oscillator strength (0.668 for the 780.2 nm line), and ϵ is the geometrical path length difference between the two arms. Unlike the hook method, where the goal is to find the two, opposite inflection points due to the anomalous dispersion, we gain more precise information by following the analysis of Hill (1986). Note that the inexpensive diode laser need not be stabilized to give results competitive with the traditional incandescent lamp and monochromator.

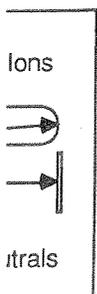
The complete experiment requires analysis of the neutral Kr produced to see what fraction is in a metastable state. Three options are available. The first is to reionize the metastable component using tuned lasers (to bring the atoms to a Rydberg state near the continuum) and electric field plates (to affect the final ionization). Fig. 5 shows this arrangement, with the first set of field plates removing from the beam any ions remaining after the Rb cell, and the second set of field plates both ionizing and deflecting those atoms excited by the lasers. In Fig. 6, we show the candidate transitions. The 811.5 nm laser would be a diode laser; the others would be CW pumped dye lasers.

The other two options are to look at resonance fluorescence after the first set of field plates in Fig. 5. In one arrangement, the $5s [3/2]^o (J=2)$ level would be pumped by the 811.5 nm laser to $5p [5/2] (J=3)$, and scattered light at the same frequency would be

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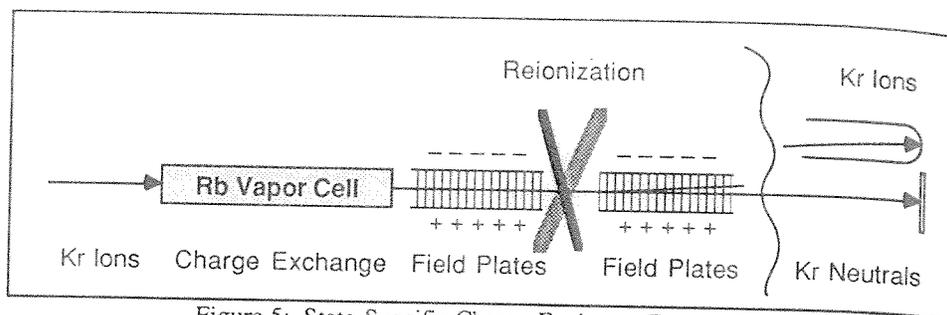


Figure 5: State-Specific Charge-Exchange Experiment

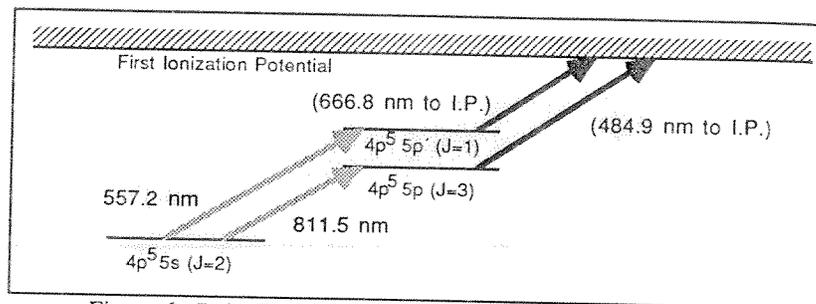


Figure 6: Reionization Pathways from the J=2 Metastable Level

detected. This is the same two-level system available for the "photon burst" detection scheme. (Photon burst techniques are discussed elsewhere in this issue.) Alternatively, the $5s [3/2]^{\circ} (J=2)$ level could be pumped by a 810.7 nm laser to the $5p [5/2] (J=2)$ level. From there, the atom would decay back to the ground state in a two-step cascade, first to the $5s [3/2]^{\circ} (J=1)$ level (emitting 877.9 nm) and then to ground state (emitting 123.5 nm). According to the calculations of Aymar and Coulombe (1978), the branching ratio for this cascade is highly favorable, with only 0.2% of the population lost, due to a transition from $5p [5/2] (J=2)$ to $5s' [1/2]^{\circ} (J=1)$. Detection of ultraviolet photons at 123.5 nm would signify creation of metastable ions.

We have special interest in this cascade process for another reason. At NBS, calibration of visible light irradiance traces back to blackbody radiation at the temperature of freezing (melting) gold. Calibration of UV irradiance traces back to synchrotron radiation. Since this cascade produces equal flux in the two spectral regions, it holds promise as a means of intercomparing the two calibration chains.

4. REFERENCES

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†Present Address: State University of New York, Buffalo, NY 14214