State Selective Charge Transfer Cross Sections for Na^+ with Excited Rubidium: A Unique Diagnostic of the Population Dynamics of a Magneto-Optical Trap

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It is shown how the newly developed technique of magneto-optical-trap recoil-ion momentum spectroscopy can be used to measure the temporal evolution of excited state fraction in such a trap. In this case, the fraction of atoms in a $5p$ state is measured. The technique can be generalized to allow the measurement of more complicated systems, e.g., a Rb sample having a mixture of $5s$, $5p$, $4d$, and Rydberg states.

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In the past several years the magneto-optical trap (MOT) has become an enabling technology for many areas of atomic, molecular, and optical physics. Examples include Bose-Einstein condensate and degenerate Fermi gas formation (and all the accompanying processes studied therein), photoassociation, slowing of the speed of light, electromagnetically induced transparency, high resolution spectroscopy, etc. In many of these studies, knowledge of population dynamics can provide greatly needed insight [1,2]. However, reliable measurements of excited state populations, either dynamic or steady state, are often difficult [2,3]. The determination of the relative populations in a laser-excited system generally relies on some model-dependent measurement [3,4] and can often become the dominant source of uncertainty [5] in the understanding of the process under study. In addition, if the excitation of the trapped atoms is not steady state, then the level populations are dynamic, requiring more complicated models and even greater uncertainty. Often, lasers are used to probe the system under study. This can have the unfortunate side effect of modifying the very populations one wishes to measure.

A specific case in which knowledge of the excited state fraction is critical is the measurement of charge transfer cross sections from excited targets. Because measured charge transfer rates are proportional to the product of the cross section for transfer from some state and the population of that state, one must have an independent measurement of the relative populations in order to obtain the cross sections.

We report here a new method for the simultaneous measurement of excited state fractions and capture cross sections. The conditions essential for the success of this method are that the relative capture rates from and into the various channels can be distinguished, and that the target is cold. Both conditions are met by magneto-optical-trap recoil-ion momentum spectroscopy (MOTRIMS) which is the focus of this work. The method can be generalized to an arbitrary number of excited levels in the target, the only limitation being the Q-value resolution of the MOTRIMS technique. The new method is independent of the mechanism by which the target has been excited and is essentially nonintrusive [6] which makes it ideal as a probe of physical processes in MOTs which result in excited states. Thus the method described in this work may be considered as a powerful tool for charge transfer measurements, or as an end in itself in which charge transfer is used to probe population dynamics in a MOT.

The target under investigation in this Letter is rubidium which is cooled and trapped using the $5s$-$5p$ cycling transition. The projectile is 7 keV Na^+. As an elementary example of the power of the method when used as a probe, we present measurements of the time evolution of the relative populations of the $5s$ and $5p$ states in the MOT as the ''repumping'' laser is chopped on and off.

MOTRIMS [7–9] is the principal tool used in these measurements. An outgrowth of the more general RIMS (recoil-ion momentum spectroscopy) [10–13] method, MOTRIMS enables higher resolution Q-value and scattering angle measurements in ionizing ion-atom or photon-atom collisions. A detailed description of the MOTRIMS apparatus used in the present study will be published separately [14]. Briefly, the TRIMS (target recoil-ion momentum spectroscopy) method consists of electrostatically extracting recoil ions created in ionizing collisions and measuring their 3-dimensional momentum vector through time-of-flight (TOF) and 2-dimensional position-sensitive detection (2D-PSD) techniques. Q values, the differences between initial and final electron binding energies, and projectile scattering angles are determined from the measured momentum components.

Of paramount importance in this technique is that the thermal momentum distribution of the target be small compared to the momentum transferred to it in the collision. Use of a MOT offers several advantages over the gas jet target typically used in TRIMS. The principal one exploited in this work is that these target species are, of necessity, readily excited by lasers, allowing for collisions studies on excited as well as ground state targets.

In the MOT used in this setup, ^{87}Rb atoms are trapped and cooled using 20 mW from a 780 nm diode laser, split

into three pairs of counterpropagating 5 mm diameter beams, which are detuned by approximately $2\pi \times 12$ MHz (two linewidths) from the $5s^2S_{1/2}$, $F = 2$-$5p^2P_{3/2}$, $F = 3$ transition. A second diode laser of the same power is similarly split and used to "repump" on the $5s^2S_{1/2}$, $F = 1$-$5p^2P_{3/2}$, $F = 2$ transition. Typically, the MOT density and temperature were measured to be approximately 5×10^{10} cm^{-3} and 200 μK, respectively. The trapping magnetic field typically has a gradient of 5 G/cm.

The Na^+ beam, typically 150 pA, is produced in a thermionic ion source, collimated, and directed through the target. Projectiles neutralized through charge transfer are detected on a 2D-PSD, while unaffected projectile ions are electrostatically deflected into a Faraday cup. The overlap between the projectile beam and the target was not well determined, thus limiting measurements to relative cross sections. The rubidium was cooled and trapped inside a recoil-ion momentum spectrometer [10,11] whose extraction axis was oriented 3.5° with respect to the projectile axis. The recoil PSD and time-of-flight signals were sent to a list-mode data acquisition system where the three components of the recoil momentum were determined, ion by ion, and Q value and scattering angle spectra were accumulated during the course of several hours at an average rate of 20–30 counts/s. A typical Q-value spectrum for the collision system under study here is shown in Fig. 1. The points represent data, while the solid line is the sum of Gaussian fits to the individual peaks. For the collision system in use for this work, the typical recoil-ion momentum resolution is 0.03 a.u. for the $p_\parallel$ (along the projectile axis) and 0.35 a.u. for $p_\perp$ (perpendicular to the projectile axis). These are limited by the timing and spatial resolution of the detector. For this collision system, $\Delta p_\parallel = 0.03$ a.u. corresponds to $\Delta Q = 110$ meV.

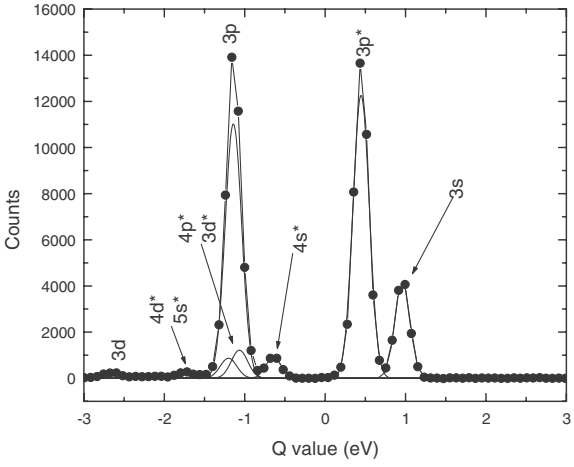

FIG. 1. Q-value plot for the 7 keV $Na^+ + Rb$ collision system. Both lasers driving the $5s$-$5p$ transition are on. Peak labels containing a star indicate channels where capture is from the $5p$ state. Other peaks represent capture from the ground state. The solid line is the sum of Gaussian fits to the individual peaks.

The basic use of the MOTRIMS method has already been demonstrated [7] in measurements of final and initial state dependant, differential in scattering angle, charge transfer cross sections for a rubidium target in a mixture of $5s$ and $5p$ states. In that measurement, a more traditional method [3] was used to determine the excited state fraction. Here we introduce a slight modification of the basic MOTRIMS technique, which allows the direct measurement of both the excited state fraction and the σ_p/σ_s ratio. The key notion is that the high resolution Q-value measurement allows one to determine which capture events come from the ground state and which come from the excited state. Thus, by chopping the trapping and repump lasers and comparing the change in charge transfer rates from the ground and excited states for lasers on and lasers off, one can determine the excited state fraction and, independently, the relative capture cross section from both states. If A_i refers to the area under a Q-value peak corresponding to charge transfer from the target's ith initial state whose relative population is given by n_i, to a particular final state, then

$$A_s \propto \sigma_s n_s, \tag{1a}$$

$$A_p \propto \sigma_p n_p, \tag{1b}$$

where the constant of proportionality contains acquisition time and geometric factors. With a high enough chopping frequency (in the case of this work, greater than 10 kHz)

$$n_s + n_p = \text{const}, \tag{2}$$

and therefore

$$\Delta A_s \propto \sigma_s \Delta n_s, \tag{3a}$$

$$\Delta A_p \propto \sigma_p \Delta n_p, \tag{3b}$$

$$\Delta n_s + \Delta n_p = 0, \tag{3c}$$

where Δn_i refers to changes in the ith population as the trapping (and/or repumping) laser goes from the on condition to the off condition. Note that it is the low temperature of the target which allows Eqs. (2) and (3c) to be satisfied at relatively low chopping frequencies.

Taking the ratio of Eq. (3b) to Eq. (3a), and using Eq. (3c) we obtain

$$\frac{\Delta A_p}{\Delta A_s} = \frac{\sigma_p}{\sigma_s}\frac{\Delta n_p}{\Delta n_s} = -\frac{\sigma_p}{\sigma_s}. \tag{4}$$

Then, using this with Eqs. (1) and (2),

$$\frac{n_p}{n_s} = \frac{\sigma_s}{\sigma_p}\frac{A_p}{A_s} = -\frac{A_p}{A_s}\frac{\Delta A_s}{\Delta A_p}. \tag{5}$$

In order to measure ΔA_i, an acousto-optic modulator (AOM) was used to chop both the trapping and repump laser beams at 50 kHz, with an on-time duty cycle of 75%. A time-to-amplitude converter (TAC) was keyed to the

AOM, and the TAC output was sent to the analog-to-digital converter (ADC) of the data acquisition system to provide a time signal, related to the laser status at the time of the collision. Thus, every time we begin a laser on/off cycle followed by a capture event, we add one count to Fig. 2. A plot of TAC output versus Q value is shown in Fig. 2. One can readily set a gate on selected individual capture channels in this figure and directly measure A_i and ΔA_i. The relative populations can then be determined using Eqs. (4) and (5). Here, we use the cross section ratio for the Rb($5p$)-Na($3p$) channel to the Rb($5s$)-Na($3s$), and then use the entire Q-value spectrum to convert from this ratio of partial cross sections to a ratio of total cross sections.

Cross section measurement is not the focus of this paper. However, as a demonstration of the validity of this technique, a series of measurements was made in which the excited state fraction in the MOT was varied by changing the detuning of the trapping laser. (Note that this also changes many MOT parameters, including trapped atom number and temperature.) In each of these measurements the ratios of the capture cross sections and the excited state fractions were independently determined. The results are shown [15] in Fig. 3. The error in the measurements is dominated by counting statistics. The figure shows that within experimental uncertainty, the cross section ratio is a constant, independent of excited state fraction. (The measured cross section ratio of 2.75 is completely consistent with the coupled channel calculation [16].) Once again, however, the emphasis of this work is on the measurement of excited state fractions. Figure 3 shows that with the MOTRIMS technique one can readily measure even small variations in the excited state fraction. If, instead, fluorescence was used to try to measure the fraction, one would have to somehow measure and correct for the varying total MOT number. With the methodology described here, this is clearly not necessary.

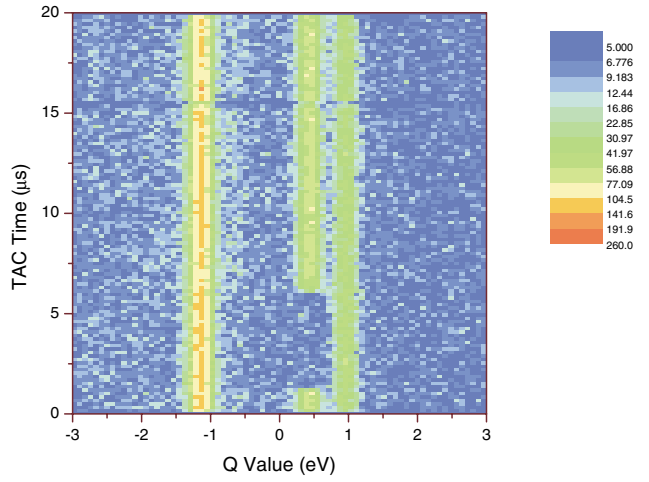

FIG. 2 (color). TAC signal versus Q value. The vertical lines can be compared with the peaks of Fig. 1. Broken lines correspond to capture from excited states.

As a demonstration of the versatility of the technique for studying MOT dynamics on a long time scale, the AOM was left on (i.e., it passes both the trapping and repump beams) and only the repump laser beam was mechanically chopped at 200 Hz with an on-time duty cycle of 75%. In order to measure the time dependence of the $5s$ and $5p$ populations, a triangle-wave signal, synchronous with the mechanical chopper, was sent to the ADC of the data acquisition computer, in place of the TAC signal. Q-value spectra were then sorted by triangle-wave voltage and phase, and Eq. (5) was used to obtain the temporal evolution of the $5s$ and $5p$ states of the target. This can be done now that σ_p/σ_s has been measured. In the results shown in Fig. 4, one can readily see the decay of the $5p$ population, resulting from optical pumping to the $F = 1$ of the ground state.

The technique used to determine the charge transfer cross sections and relative populations is completely general, and could in principle be used to determine cross sections and populations for a system containing N levels. Equations (4) and (5) can be generalized to expressions for the kth level ($2 \leq k \leq N$):

$$\frac{\sigma_k}{\sigma_1} = -\Delta A_k \left(\sum_{i=1}^{k-1} \Delta A_i \frac{\sigma_1}{\sigma_i} \right)^{-1}, \tag{6a}$$

$$\frac{n_k}{n_1} = \frac{\sigma_1}{\sigma_k} \frac{A_k}{A_1}, \tag{6b}$$

where the subscript "1" indicates the lowest level, and the Δ refers to changes as the laser driving the kth level is chopped on and off. The terms σ_i/σ_1 are sequentially determined for all $i \leq k$ by iteratively using Eq. (6a). Once all of the relative cross sections are determined, Eq. (6b) can be used to determine the temporal evolution of each of the N levels.

Using the MOTRIMS technique, resolution in the time evolution of the populations in a system is limited only by the resolution of our time-of-flight measurements, here, a

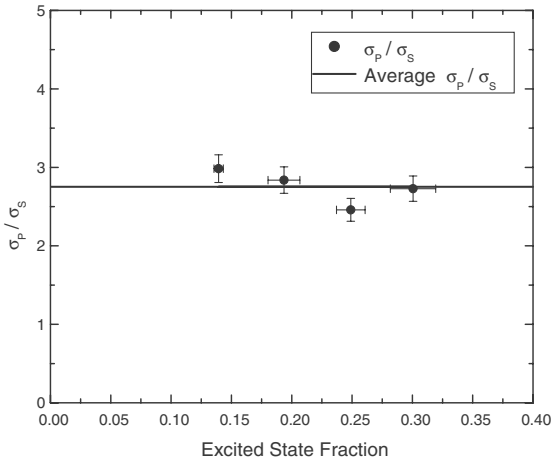

FIG. 3. Ratio of capture cross sections versus measured excited state fraction. Error bars are absolute.

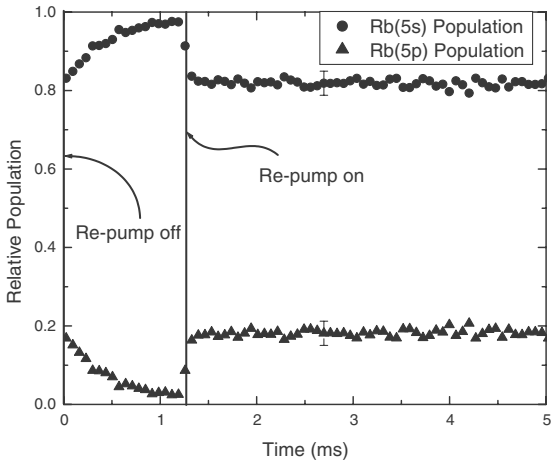

FIG. 4. Time dependence of relative Rb(5s) and Rb(5p) populations. At $t = 0$, the light from the "repump" laser was blocked. The vertical line near 1.2 ms indicates at which point the beam was turned back on. Representative error bars are absolute; relative error is smaller than the size of the points.

few nsec [17]. It is apparent that this technique is more than just a tool for the determination of charge transfer cross sections, but rather is a very general and powerful method for studying the temporal evolution of level populations in a MOT under the influence of some perturbation [18]. Because of the small capture cross section coupled with the high detection efficiency, the use of the ion beam as a probe has virtually no effect on the MOT. Example systems which one might study include the formation of cold molecules [19] or cold Rydberg plasmas [20,21]. One could as well study the dynamics of the number of atoms in the MOT being dressed by a fast laser pulse, or the influence on populations by the "coupling" laser in an electromagnetically induced transparency experiment [22].

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