

## Kinematically Complete Charge Exchange Experiment in the $\text{Cs}^+ + \text{Rb}$ Collision System Using a MOT Target

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Charge exchange is examined with unprecedented precision using the newly developed magneto-optical trap-target recoil ion momentum spectroscopy (MOTRIMS) technique. Initial and final state selective, charge exchange cross sections are obtained for 6 keV  $\text{Cs}^+$  colliding with rubidium in  $5s$  and  $5p$  states. For each charge transfer channel, cross sections differential in scattering angle are measured. These data are used to overturn previous long-standing conjecture as to the origin of oscillations seen in total charge exchange cross section measurements, and compare well with an enhanced Demkov model calculation.

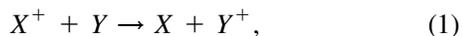
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In slow ion-atom collisions charge exchange is the dominant process. This mechanism thus plays an important role in any natural plasma environment, such as the upper atmosphere, the solar corona, and interstellar media, or in man-made plasmas such as those produced in tokamaks and in cold plasma processing systems [1–4]. Ideally, one should be able to accurately predict the outcome of binary collisions between constituent species, and then use these predictions in more comprehensive theoretical treatments which model realistic, complicated environments. However, although the general nature of single electron transfer in low energy collisions is fairly well understood, some technically difficult problems remain. For example, in coupled channel calculations which make use of a model potential for the core, it is challenging to correctly take into account the effects of core polarization in low velocity charge exchange collisions.

In order to provide the most stringent test of theory in the binary collisions, one would therefore like to have experimental results which are differential in as many parameters as possible. In particular, a scattering experiment which is differential in initial state, final state, energy, and scattering angle, is termed a “complete experiment” [5], and is most helpful in providing a guidepost for the theorist developing these models.

A case in point is the charge exchange reaction given by



where  $X$  and  $Y$  are alkali atoms. It is particularly important that theoretical models be successful with alkali-alkali collision systems, since by virtue of having precisely one active electron, these systems represent one of the simplest class of collision partners. Furthermore, unlike atomic hydrogen, the alkalis are relatively easy to produce in the laboratory, thereby providing a means for testing theoretical models.

In the late 1960’s Perel and co-workers, using then state-of-the-art techniques, were able to detect distinct oscilla-

tions in the total charge transfer cross sections as a function of collision energy in alkali ion-alkali atom systems [6]. It was conjectured at that time that the oscillations were caused by the projectile ion polarizing the target atom, thereby putting it into a superposition of  $s$  and  $p$  states. The ensuing charge transfer process could then follow along two distinct paths, resulting in the observed interference pattern. Theoretical treatments of the collision [7,8] neither confirmed nor disputed these conclusions, and the hypothesis has remained unchallenged for roughly thirty-five years, at least partly because no experimental method existed which could test it. In this paper we demonstrate a new high resolution technique, MOTRIMS, and implement it to test this hypothesis.

Specifically, we present the results of an investigation of the particular collision system  $\text{Cs}^+ + \text{Rb}$  using the newly developed MOTRIMS (magneto-optical trap-target recoil ion momentum spectroscopy) system. MOTRIMS is the latest development [9–11] in the more general COLTRIMS (cold TRIMS) experimental method [12,13]. Subject only to resolution and target species constraints, the TRIMS technique allows one to perform a kinematically complete charge exchange experiment. In the MOTRIMS technique described here, these constraints are pushed back to achieve unprecedented resolution and extension to target systems not possible in the usual COLTRIMS setup.

The TRIMS method consists of electrostatically extracting recoil ions created in ionizing collisions, measuring their three-dimensional momentum vector through time-of-flight (TOF) and two-dimensional position sensitive detection (2D-PSD) techniques, and determining  $Q$  values and scattering angles from this measured momentum. 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. This is typically accomplished by precooling the target, and then allowing it to undergo supersonic expansion. The technique is a powerful one and a tremendous amount of collisions physics has been understood through its use [12,13].

However, even by cooling helium targets down to near-liquid helium temperatures, followed by supersonic expansion, the resolution of the technique is still limited by target temperature. In addition, conventional COLTRIMS is limited to target species which can be precooled and then expanded to achieve very low temperatures. This excludes most targets, one of which, the alkalis, are of particular interest because they represent single active electron systems. The alkalis have heretofore not been subjected to this powerful experimental probe.

A simplified schematic of the experimental setup is shown in Fig. 1. In the target region,  $^{87}\text{Rb}$  atoms are trapped and cooled in a magneto-optical trap (MOT) using a diode laser, split into three pairs of counterpropagating beams which operate on the  $5s^2S_{1/2}, F = 2-5p^2P_{3/2}, F = 3$  transition. A second diode laser is used to “re-pump” on the  $5s^2S_{1/2}, F = 1-5p^2P_{3/2}, F = 2$  transition. The rubidium target density was measured to be  $(4 \pm 3) \times 10^{10}/\text{cm}^3$ ; its diameter was  $0.5 \pm 0.1$  mm; and its temperature was  $130 \pm 100$   $\mu\text{K}$ . The overlap between the projectile beam and the target was not well determined, thus limiting the measurements to relative cross sections. The trapping magnetic field had a gradient of approximately 5 G/cm. The rubidium was cooled and trapped inside a recoil ion momentum spectrometer [12,13] whose extraction axis was oriented  $3.5^\circ$  with respect to the projectile axis, as shown in Fig. 1. The three components of the recoil momentum are determined, ion by ion, through their time of flight, and their positions on a 2D-PSD. The projectiles continue past the recoil detector, and are charge analyzed, with the neutral cesium striking a second 2D-PSD, and the  $\text{Cs}^+$  being collected in a Faraday cup. The component of recoil momentum parallel to the projectile axis,  $p_{\parallel}$ , is directly related to the  $Q$  value [12,13], while the components of the momentum perpendicular to the projectile axis,  $p_{\perp}$ , are directly related to the scattering angle. Although transverse extraction is more common, our spectrometer was designed for longitudinal extraction [14,15] so as to optimize the resolution of  $p_{\parallel}$ , and thus of the  $Q$  value. As is the case for most COLTRIMS systems,

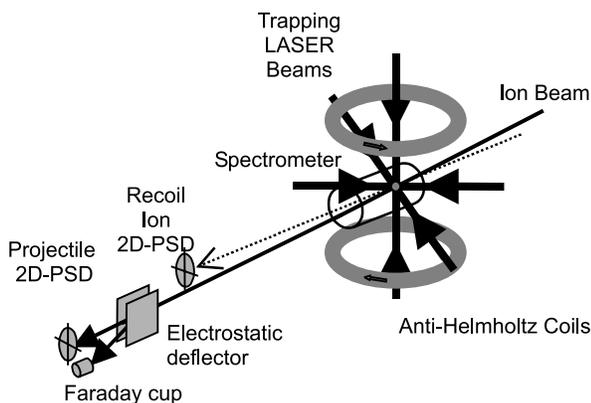


FIG. 1. Simplified schematic of the experimental apparatus.

the component of the three-dimensional momentum vector derived from the time of flight tends to have better resolution than the components measured from the 2D-PSD. For the experiment described here the TOF was approximately 70  $\mu\text{sec}$ , and was measured with a precision of about 2 nsec. It is not clear what is determining the resolution of the TOF measurement, but we suspect that a slight energy spread in the projectile beam (of the order of 1 eV out of 6 keV is sufficient) may be setting the limit. The small angle between the beam axis and extraction field has negligible effect on the timing or spatial resolution.

Figure 2 shows a typical  $Q$ -value spectrum for this collision system. The data are shown as points, and the lines are the results of a model calculation convoluted with Gaussians, each of width 96 meV, the instrumental resolution. This corresponds to a recoil ion momentum resolution of about 0.08 a.u. [16] (or a velocity resolution of  $0.5 \times 10^{-6}$  a.u.  $\sim 1$  m/s). Thus, for equivalent target mass, the MOTRIMS method represents about an order of magnitude improvement in momentum resolution over conventional COLTRIMS. Without the unprecedented resolution of this MOTRIMS technique, initial and final state selectivity would not be possible for this collision system. The peaks are labeled by the final state in  $^{133}\text{Cs}$  occupied by the transferred electron. Because this is a  $Q$  value, or energy defect spectrum, one can readily distinguish between transfer from ground state or excited rubidium [17]. Note that nearly all of the charge transfer from ground state rubidium goes to the ground state of cesium, though a small amount may be captured into the

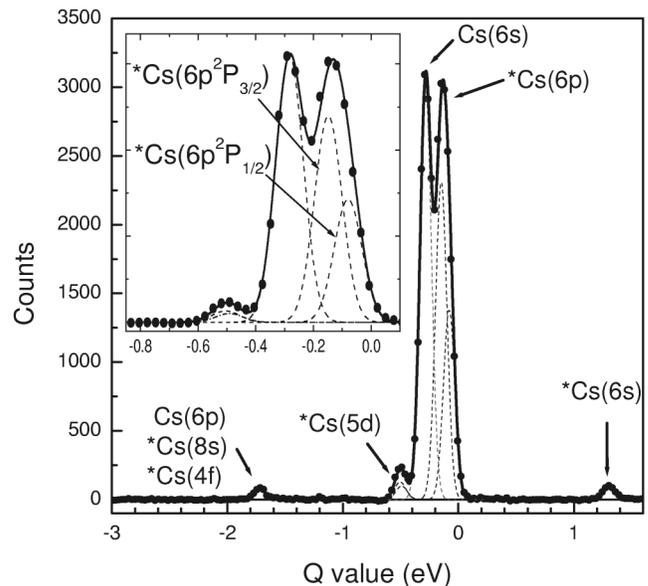


FIG. 2. Counts versus  $Q$  value, or energy defect. The labels identify the final state in cesium; “\*” on the labels indicates charge exchange from  $\text{Rb}(5p)$ . The solid points are data; the inset shows the theoretical predictions of individual channel cross sections (dashed lines) and the sum of all the channels (solid line).

Cs(6p). By contrast, electrons from excited rubidium are captured into several states in cesium. This is expected since in the latter case there are more low  $Q$ -value channels accessible.

Figure 3 shows the charge transfer cross section, differential in scattering angle, for each of the major peaks of Fig. 2. The resolution in scattering angle, essentially limited by the resolution of the position sensitive detector, is about  $30 \mu\text{rad}$ . Distortions due to the magnetic field gradient, consistent with ion trajectory calculations, lead to a rotation and “squeezing” of the recoil ion spot. This distortion is readily corrected in software and does not appear to degrade the resolution in scattering angle. Structure is clearly seen in the Rb(5s)-Cs(6s) case and, to a lesser extent, in the Rb(5p)-Cs(6p) case as well. For both of these cases, oscillations arise from the Hankel transform [18] of the impact parameter dependent scattering amplitude. In the “s-s” channel,  $m_i = m_f = 0$  which means exactly one term (and therefore one phase) exists in the transform. However, in the “p-p” channel,  $m_i = 0, \pm 1$  and  $m_f = 0, \pm 1$ . Therefore, the different phases of each of these terms may cause some of the structure to be washed out. (Here  $m_i$  and  $m_f$  refer to the initial and final magnetic quantum numbers, respectively.) For the other transitions, better statistics may reveal some structure, but at a greatly reduced level as compared to the Rb(5s)-Cs(6s) and Rb(5p)-Cs(6p) cases. These cross sections are essentially quadruply differential (scattering angle, initial state, final state, and collision energy) and therefore serve as extremely severe tests of theory.

The  $Q$ -value results of these experiments show conclusively that the structure observed by Perel [6] cannot possibly be due to polarization effects in the incoming

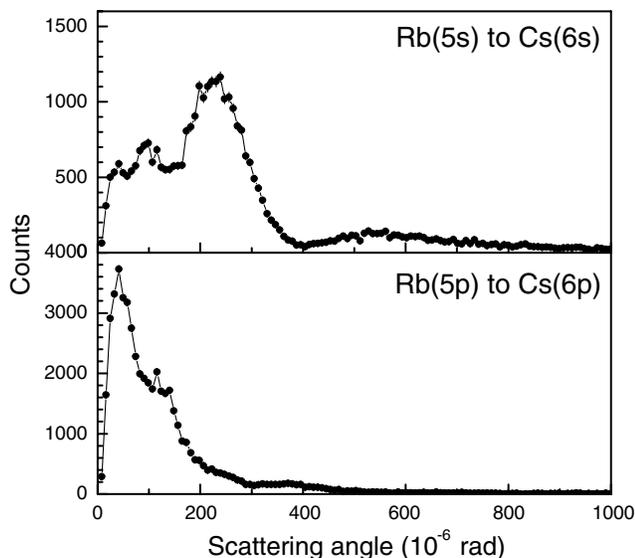


FIG. 3. Counts versus scattering angle for the two dominant channels shown in Fig. 2. The resolution is approximately  $30 \mu\text{rad}$  (four points) and the error bars are statistical. The solid line simply connects the points to guide the eye.

channel causing interferences. The evidence is twofold: First of all, we can see from the laser-excited rubidium lines in Fig. 2 that rubidium in the 5p state transfers predominantly to Cs(6p), with some small fraction going to Cs(5d), and an even smaller fraction going to Cs(6s). Secondly, “direct” charge exchange from ground state rubidium would have precisely the same  $Q$  value as “polarization” charge exchange (the mechanism proposed by Perel *et al.*). Therefore, if Perel’s hypothesis were correct, Fig. 2 should show a large peak at a  $Q$  value corresponding to the Rb(5s)-Cs(6p) transition, whereas the actual peak at that  $Q$  value is extremely small. Furthermore, we can say that the amplitude of the relatively small Rb(5p)-Cs(6s) peak represents an upper limit to the contribution of polarization charge exchange to the large Rb(5s)-Cs(6s) peak. This upper limit does not seem large enough to produce the interference structure seen in Perel’s measurements. Thus, a different mechanism must be responsible for the oscillations reported by Perel.

In order to theoretically compare the cross sections for the different charge exchange channels with each other, the explicit impact-parameter dependence of the transition probability was derived [19] within the Demkov model [8]. In this model, each transition is treated as a two-coupled-states problem, neglecting the effect of the other final states involved. The coupling between the states is assumed to be  $H_{12} \simeq \beta \exp[-\sqrt{2I} R(t)]$ . Thus by substituting  $R(t) = \sqrt{(vt)^2 + b^2}$ , where  $b$  is the impact parameter, the  $b$  dependence is explicitly introduced, leading to the transition probability

$$P(b) = \text{sech}^2(\pi\Delta/2\gamma) \sin^2[2\beta K_0(2Ib^2)/v\sqrt{2I}], \quad (2)$$

where  $\gamma = v\sqrt{2I}\sqrt{1 - 2Ib^2/\ln^2\Delta/\beta}$ ,  $\Delta = |I_1 - I_2|$ ,  $I_1$  and  $I_2$  are the ionization potentials of the initial and final states,  $I$  is the minimum of  $I_1$  and  $I_2$ ,  $v$  is the collision velocity,  $\beta \simeq I$  as suggested by Meyerhof [20],  $K_0$  is a  $K$ -Bessel function, and all values are in atomic units. The cross sections of the reaction channels for which  $H_{12} \gg \Delta$  are then evaluated by integration over  $b$ , and compared to the data after folding in the experimental resolution of 96 meV.

Shown in the inset of Fig. 2 are the theoretical predictions of individual channel cross sections (dashed lines) and the sum of all the channels (solid line). Two parameters were used to fit the results of this model calculation to experiment. These are an overall multiplicative constant (only relative cross sections were measured) and a target excitation fraction. From the fit, an excitation fraction of 15% was determined. This is well within the uncertainty of an independent measurement of the excited state fraction of  $16\% \pm 7\%$ . The modified Demkov model greatly underestimated the size of the Rb(5p)-Cs(6s) channel. This is expected since the model is valid only for  $H_{12} \gg \Delta$ . While this approximation is valid for channels having a  $Q$  value  $< \pm 0.8$  eV, it evidently fails for the larger  $Q$  values.

In this Letter we have demonstrated a new technique, MOTRIMS. With this method we extend the number and type of target atoms which can be investigated by TRIMS. Furthermore, because target temperature is no longer a limitation, significant improvements in recoil momentum resolution can be achieved. Such high resolution differential measurements, both in  $Q$  value and angle, provide benchmark data which can be used to further develop theoretical treatment. To illustrate the power of MOTRIMS, we have presented a study of an alkali collision system, namely, charge capture from Rb( $5s$ ) and Rb( $5p$ ) by 6 keV Cs<sup>+</sup> projectiles. We have shown that for ground state rubidium, virtually all charge exchange ends up in the ground state of cesium. Furthermore, because Rb( $5p$ )-Cs( $6s$ ) is negligible, we argue that the oscillations reported by Perel *et al.* were not due to target polarization as had been proposed [6]. We have introduced a modification to the Demkov model, in which impact parameter dependence is explicitly given. Calculations based on this model agree extremely well with dominant charge exchange channels, those having small  $Q$  values.

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- [1] R.R. Hodges, Jr. and E.L. Breig, *J. Geophys. Res.* **96**, 7697 (1991).
- [2] S.A. Fuselier, E.G. Shelley, B.E. Goldstein, R. Goldstein, and M. Neugebauer, *Astrophys. J.* **379**, 734 (1991).
- [3] P.C. Stancil, S. Lepp, and A. Dalgarno, *Astrophys. J.* **509**, 1 (1998).
- [4] R.C. Isler, *Plasma Phys. Controlled Fusion* **36**, 171 (1994).
- [5] B. Bederson, *Comments At. Mol. Phys.* **1**, 41 (1969); **1**, 65 (1969).
- [6] J. Perel, R.H. Vernon, and H.L. Daley, *Phys. Rev.* **138**, A937 (1965).
- [7] D. Rapp and W.E. Francis, *J. Chem. Phys.* **37**, 2631 (1962).
- [8] Yu.N. Demkov, *Sov. Phys. JETP* **18**, 138 (1964).
- [9] S. Wolf and H. Helm, *Phys. Rev. A* **56**, R4385 (1997).
- [10] S. Wolf and H. Helm, *Phys. Rev. A* **62**, 043408 (2000).
- [11] M. van der Poel, Ph.D. thesis, Niels Bohr Institute, University of Copenhagen, 2001 (unpublished).
- [12] J. Ullrich, R. Moshhammer, R. Dörner, O. Jagutzki, V. Mergel, H. Schmidt-Böcking, and L. Spielberger, *J. Phys. B* **30**, 2917 (1997).
- [13] R. Dörner, V. Mergel, O. Jagutzki, L. Spielberger, J. Ullrich, R. Moshhammer, and H. Schmidt-Böcking, *Phys. Rep.* **330**, 95 (2000).
- [14] R. Moshhammer, M. Unverzagt, W. Schmitt, J. Ullrich, and H. Schmidt-Böcking, *Nucl. Instrum. Methods Phys. Res., Sect. B* **108**, 425 (1996).
- [15] H. Kollmus, W. Schmitt, R. Moshhammer, M. Unverzagt, and J. Ullrich, *Nucl. Instrum. Methods Phys. Res., Sect. B* **124**, 377 (1997).
- [16] Using a different projectile beam, the collisions physics of which is not germane to this Letter, we have obtained a  $p_{\parallel}$  resolution of 0.03 a.u.
- [17] The trapping and repump lasers leave approximately 16%  $\pm$  7% of the rubidium target in the  $5p$  state.
- [18] W. Fritsch and C.D. Lin, *Phys. Rev. A* **54**, 4931 (1996).
- [19] Details will be presented in a future publication.
- [20] W.E. Meyerhof, *Phys. Rev. Lett.* **31**, 1341 (1973).