Q-Value Measurements in Charge-Transfer Collisions of Highly Charged Ions with Atoms by Recoil Longitudinal Momentum Spectroscopy

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We report on the first Q-value measurements in charge-transfer collisions using recoil longitudinal momentum spectroscopy. This method is not limited to relatively low beam energies and is easily adaptable to captures involving any number of transferred electrons. A very monoenergetic beam is not necessary. For a 50-keV Ar^{15+} on Ar collision system, Q values corresponding to single through quintuple electron capture were measured and found to be in good agreement with the predictions of the molecular classical overbarrier model.

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When a multiply charged ion moving at a velocity below that of typical outer shell atomic electrons encounters a neutral target, the dominant electron removal process is electron capture. The change in electronic energy, or Q value, for such a process is a direct measure of the distribution of final states populated on the projectile, which is in turn one of the most crucial tests of any theoretical description of the process. Direct measurement of the energy gain of the projectile has often been used to determine this final-state distribution [1-16]. Since the fractional energy resolution practically achievable rarely exceeds 10^{-3}, this technique has usually been limited to projectiles of 10 keV and less, and to single and double capture. For higher energies, measurement of Q through direct energy change of the projectile becomes increasingly more difficult [17]. It has been pointed out previously that recoil ion longitudinal momentum can yield direct information on the Q value and electron mass transfer in fast-ion–atom collisions [18]. In this work we report the first experimental use of recoil longitudinal momentum spectroscopy to obtain Q values for capture. The technique is not limited to collision energies below a few keV, and can be used in situations for which the beam energy is not very well determined. This technique is readily applicable to any charge-transfer number, and is not affected by the kinematic broadening due to autoionization of the projectile following the collision. We apply the method to measure Q values for up to five-electron transfer for Ar^{15+} ions on Ar.

For collisions involving i-electron capture, conservation of both energy and momentum results in a simple relation between the Q value of the collision and the momentum transfer to the recoil given by (in a.u.)

$$Q=Q_0+Q'\approx -\left[P_{\parallel}v+\frac{iv^2}{2}\right]+\frac{M_1+M_2}{2M_1M_2}(P_{\parallel}^2+P_{\perp}^2)\,, \tag{1}$$

where v is the projectile velocity, $P_{\parallel}$ and $P_{\perp}$ are the longitudinal and transverse (relative to the beam direction) components of the momentum transfer to the recoil, and M_1 and M_2 are the projectile and recoil masses, respectively. The term $iv^2/2$ appears due to the fact that the captured electrons are moving with the projectile at velocity v just after the collision. We will show later that Q' is much smaller than Q_0 and to a very good approximation

$$Q\approx Q_0=-(P_{\parallel}v+iv^2/2) \tag{2}$$

such that it is sufficient to measure $P_{\parallel}$ to obtain Q values.

We wish to emphasize that this technique applies only to pure electron capture collisions, and breaks down when electrons are directly ionized during the collision. By using the coincidence detection of charge state analyzed projectile and recoil, employed here, one can always isolate experimentally those channels for which no electrons are left in the continuum. Since these channels may be weakly populated, as is the case here, it is important to realize that most continuum electrons generated in capture by highly charged ions below 1 a.u in velocity result from post-collision autoionization of the projectile following capture, for which Eq. (2) remains valid. Direct target ionization during the collision, which renders Eq. (2) invalid, becomes an important process at high collision velocities, typically above 1 a.u. For example, experimental investigations have shown that the ratio of single ionization to single capture cross sections is less than 2% for Ar^{15+} and Ar^{16+} ($v=0.2$-1.6 a.u.) on He [19], and less than 5% for Ar^{17+} ($v=1.25$ a.u.) on Ar [20]. Pure ionization has been observed in Xe^{q+} ($q=15,20,25,30,35$; $v\approx 0.1$-0.2 a.u.) on Xe [21], for which the ratio of the single ionization to single capture cross section amounted to a few percent. We therefore believe that this technique can be extended to collision velocities up to about 1 a.u. where direct ionization accompanying capture is at most a few percent of the pure capture processes, and hence introduces only small errors in the measured Q values. It is worth mentioning that the deviation from Eq. (2) at higher collision velocities when ionization is substantially enhanced is in itself another interesting topic in fast-ion–atom collisions [22].

Inspection of Eq. (2) shows that the beam energy spread is not a major factor in determining the Q-value

resolution. For example, the beam used in this experiment had a modest $\Delta E/E$ of about 1/325. However, it only gives rise to an uncertainty in Q of about 0.6% in the case of quintuple electron capture for which the longitudinal momentum transfer was about 23 a.u. This allows for the extension of Q-value measurements in charge-transfer collisions to beam velocities (≈ 1 a.u.) that are not accessible to energy gain spectroscopy. The resolution of this technique is limited by the thermal longitudinal momentum spread (along the beam direction) of the target atoms. In this experiment, it was about 8 a.u. in momentum which, at the beam velocity of 0.22 a.u., corresponds to 1.76 a.u. in Q, or 48 eV. With the present resolution, it is only possible to obtain average Q values. Improvement on the resolution will be discussed later.

The experimental setup is shown in Fig. 1(a). The 50-keV Ar^{15+} beam was extracted from the Kansas State University Cryogenic Electron-Beam Ion Source (KSU CRYEBIS) [23], and directed to the collision chamber. Collimation was provided by a four-jaw slit (0.8 mm width) and the collision chamber entrance aperture (0.4 mm diameter) which were separated by about 3.5 m and thus limiting the beam divergence to less than 0.01°. The collision chamber exit (3.2 mm diameter) allowed for scattering angles up to 33 mrad. The Ar gas target was furnished by a multichannel array molecular jet, and the gas flow was adjusted to minimize double collisions such that only about 2% of the projectiles that changed their charge state were observed to have undergone double encounters. After the collisions, the recoil ions were extracted transverse to the beam direction by a uniform electric field (≈ 10 V/cm) and detected by a two-dimensional position-sensitive channel-plate detector which had a 40-mm active diameter and a resolution of 0.11 mm. A coincident time-of-flight technique was used to determine the recoil charge states. A parallel-plate electrostatic deflector separated the final projectile charge states which were then detected by another two-dimensional position-sensitive channel-plate detector located 1.2 m downstream, which also had a 40-mm active diameter but a resolution of 0.5 mm. The beam divergence, the flight path to the detector, and the detector resolution gave an angular resolution of about 0.45 mrad. Figure 1(b) shows the coordinate system adopted in the analysis.

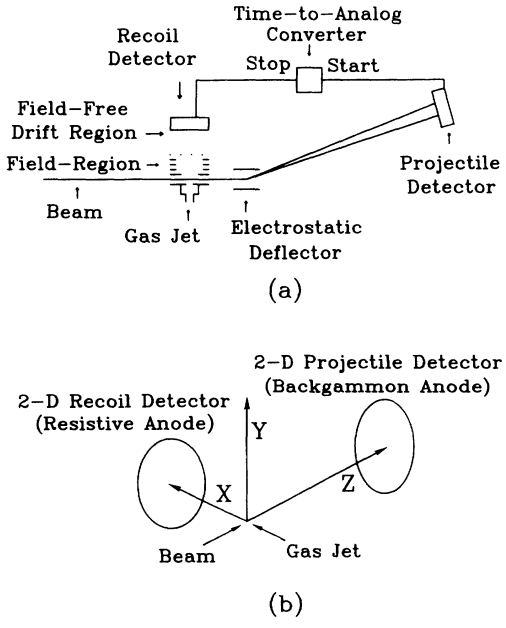

FIG. 1. Schematic of (a) the experimental setup, and (b) the coordinate system used in the analysis.

To obtain Q values, only the Z component of the recoil position vector and the time of flight are needed. Figure 2 shows the two-dimensional recoil position spectra corresponding to recoil charge states i and projectile charge changes k as well as their projections onto the beam axis (Z axis). The centroids of the projections can be used to determine the average values of the quantities of interest. To determine the average shift in Z due to the longitudinal momentum transfer, it is important to determine the zero-point (Z_0) corresponding to zero longitudinal momentum transfer. This was accomplished by applying

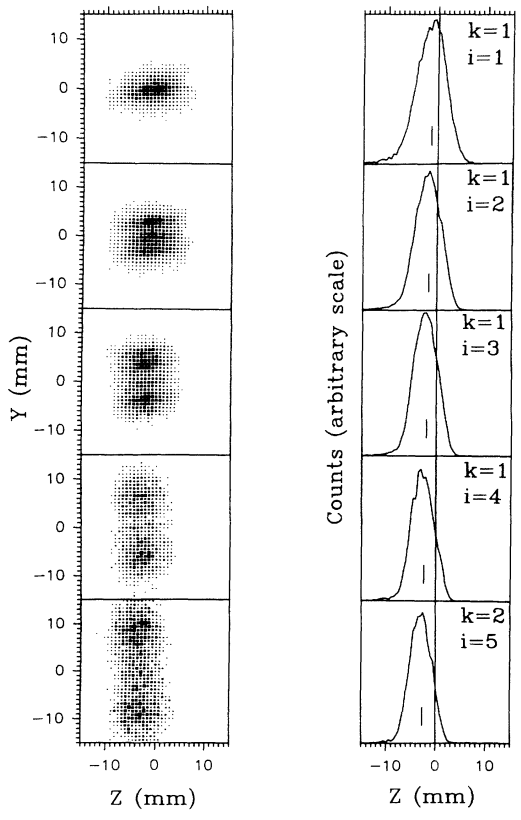

FIG. 2. Two-dimensional recoil position spectra for the different combinations of projectile charge change k and recoil charge state i, and their projections onto the Z axis. The centroids of the projections are indicated by the vertical bars.

different extraction fields (between 5 and 15 V/cm) that resulted in different times of flight and different Z positions for each recoil charge state. With the longitudinal momentum transfer being independent of the extraction field, Z_0 could be determined using the simple kinematic equation

$$Z_i = Z_0 + v_{i\parallel} t_i , \tag{3}$$

where $v_{i\parallel}$ and t_i are the longitudinal velocity and time of flight of the i-times ionized recoil, respectively. For this purpose we chose recoil charge state $i=2$ and performed a linear fit according to Eq. (3). The uncertainty in Z_0 is dominated by the uncertainty in alignment and the uncertainty in the intercept of the linear fit. All other uncertainties were found to be negligible.

The centroids of the Z positions are indicated by the vertical bars in Fig. 2. We observe that these centroids have negative values consistent with what one would expect in exoergic collisions where the recoils are thrown backwards. Having obtained the necessary position and time information, the longitudinal momentum transfer was obtained from

$$P_{i\parallel} = M_2 Z_i / t_i . \tag{4}$$

The corresponding Q_0 values were then obtained using Eq. (2) and are listed in Table I. On the average the Q value was about 25 eV for each captured electron. While recoil charge states higher than $i=5$ have been observed, they had substantially lower statistics to be considered. Neglecting Q' induces small shifts towards smaller Q values reminiscent of the kinematic shift often encountered in energy gain spectroscopy [15]. There are two ways to obtain $P_\perp$ and therefore the magnitudes of the shifts. One way is to reconstruct the recoil momentum vector from the position and time information, a technique used by Frohne *et al.* [22] in their recoil momentum spectroscopy studies at high collision energies, and hence obtain $P_\perp$. Another, which we employed, is to use the projectile angular distributions to determine the average scattering angles ($\bar{\theta}$), and by conservation of the transverse component of momentum

$$P_\perp \approx P_0 \bar{\theta} , \tag{5}$$

where P_0 is the incident projectile momentum. The average scattering angles were obtained from the angular distributions and the shifts Q' were then determined. Both $\bar{\theta}$ and Q' are also listed in Table I. It is obvious that the shifts are rather small and neglecting them does not lead to serious deviations from the true values. It should be noted that while this is the first time recoil momentum spectroscopy is used to obtain Q values for capture, recoil ion energy analysis has been used in the past to determine the inelastic energy loss of inner-shell excitation processes for small impact parameter collisions with singly charged projectiles [24].

TABLE I. Experimental Q_0, $\bar{\theta}$, Q', Q, and predicted MCBM Q values.

k	i	Q_0 (eV)	$\bar{\theta}$ (mrad)	Q' (eV)	Q[a] (eV)	MCBM (eV)
1	1	28.7	0.96	0.1	28.8 ± 4.3	25.3
1	2	51.5	1.50	0.3	51.8 ± 6.0	51.5
1	3	76.5	2.41	0.6	77.1 ± 7.4	78.4
1	4	106.4	4.11	1.8	108.2 ± 8.6	107.3
2	5	136.2	6.00	3.8	140.0 ± 9.6	133.7

[a]Quoted errors are 1 standard deviation.

In the absence of a rigorous quantum mechanical treatment of multiple-electron capture collisions, the classical overbarrier model [25] was extended by Bárány *et al.* [26] and Niehaus [27] to include such collisions in an effort to gain a better understanding of the physics involved. The different versions of the molecular classical overbarrier model (MCBM) have reasonably accounted for experimental measurements of cross sections, peak widths and positions of energy gain spectra, and angular distributions. We have used the model proposed by Niehaus to calculate Q values for the different (k,i) combinations that we measured. In these calculations we assumed the number of electrons that became molecular to be equal to the recoil charge state i. The predicted values together with the shift corrected Q values are also listed in Table I. The agreement between the measured Q values and the predictions of the MCBM is surprisingly very good considering the complexity of the physics involved and the simplicity of the MCBM assumptions.

Although the target thermal spread limited the resolution such that a single Z-position spectrum with well resolved peaks could not be obtained, such structure has primarily been seen previously only in the single-capture channel. For multiple capture, the high density of final states and the loss of projectile energy resolution due to the emission in flight of autoionization electrons has generally resulted in the observation of structureless peaks [14,28–30]. For such multiple-capture processes, we thus see no advantage of conventional energy gain spectroscopy over the present technique, even for low projectile energies. Indeed, our method has the advantage that the final channels are easily identified through coincident charge-state determination, allowing us to unambiguously determine the number of electrons captured initially, as given by the recoil charge state, and the number remaining on the projectile after autoionization, as given by the projectile charge state. However, we are in the process of building a cold gas jet to reduce the target longitudinal momentum spread to about 1 a.u. for Ar gas. This corresponds to a resolution in Q of about 1 a.u. (27 eV) for a 1-a.u. beam velocity and to 0.2 a.u. (5.5 eV) for a 0.2-a.u. beam velocity. While such a resolution could be matched by projectile energy gain spectroscopy at relatively low beam velocities, it is far from being matched for velocities above 0.5 a.u.

In conclusion, we have used recoil longitudinal momen-

tum spectroscopy to provide information on Q values in two-body reactions. We have applied it to a 50-keV Ar^{15+} on Ar collision system, and obtained Q values for up to quintuple electron capture collisions. The measured values compare well with the predictions of the MCBM. With a cold target gas jet the technique offers advantages over projectile translational energy spectroscopy and can be extended to include higher collision velocities and other projectile and target species.

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[1] R. Mann, C. L. Cocke, A. S. Schlachter, M. Prior, and R. Marrus, Phys. Rev. Lett. **49**, 1329 (1982).

[2] S. Ohtani, Y. Kaneko, M. Kimura, N. Kobayashi, T. Iwai, A. Matsumoto, K. Okuno, S. Takagi, H. Tawara, and S. Tsurubuchi, J. Phys. B **15**, L535 (1982).

[3] C. Schmeissner, C. L. Cocke, R. Mann, and W. Meyerhof, Phys. Rev. A **30**, 1661 (1984).

[4] B. A. Huber, H-J. Kahlert, and K. Wiesemann, J. Phys. B **17**, 2883 (1984).

[5] E. H. Nielsen, L. H. Andersen, A. Bárány, H. Cederquist, J. Heinemeier, P. Hvelplund, H. Knudsen, K. B. MacAdam, and J. Sørensen, J. Phys. B **18**, 1789 (1985).

[6] H. Cederquist, L. H. Andersen, A. Bárány, P. Hvelplund, H. Knudsen, E. H. Nielsen, J. O. K. Pedersen, and J. Sorensen, J. Phys. B **18**, 3951 (1985).

[7] J. P. Giese, C. L. Cocke, W. Waggoner, L. N. Tunnell, and S. L. Varghese, Phys. Rev. A **34**, 3770 (1986).

[8] R. W. McCullough, S. M. Wilson, and H. B. Gilbody, J. Phys. B **20**, 2031 (1987).

[9] E. Y. Kamber, C. L. Cocke, J. P. Giese, J. O. K. Pedersen, and W. Waggoner, Phys. Rev. A **36**, 5575 (1987).

[10] S. M. Wilson, R. W. McCullough, and H. B. Gilbody, J. Phys. B **21**, 1027 (1988).

[11] E. Y. Kamber, Nucl. Instrum. Methods Phys. Res., Sect. B **40/41**, 13 (1989).

[12] H. Cederquist, L. Liljeby, C. Biedermann, J. C. Levin, H. Rothard, K. O. Groeneveld, C. R. Vane, and I. A. Sellin, Phys. Rev. A **39**, 4308 (1989).

[13] C. Biedermann, H. Cederquist, L. R. Andersen, J. C. Levin, R. T. Short, S. B. Elston, J. P. Gibbons, H. Anderson, L. Liljeby, and I. A. Sellin, Phys. Rev. A **41**, 5889 (1990).

[14] P. Roncin, M. N. Gaboriaud, and M. Barat, Europhys. Lett. **16**, 551 (1991).

[15] C. L. Cocke and R. E. Olson, Phys. Rep. **205**, 153 (1991).

[16] H. Lebius and B. A. Huber, Z. Phys. D **23**, 61 (1992).

[17] R. Schuch, H. Schöne, P. D. Miller, H. F. Krause, P. F. Dittner, S. Datz, and R. E. Olson, Phys. Rev. Lett. **60**, 925 (1988).

[18] R. Dörner, J. Ullrich, O. Jagutzki, S. Lencinas, A. Gensmantel, and H. Schmidt-Böcking, in *Electronic and Atomic Collisions*, edited by W. R. MacGillivray, I. E. McCarthy, and M. C. Standage (Adam Hilger, Bristol, 1992), p. 351.

[19] W. Wu *et al.* (to be published).

[20] R. Ali *et al.* (to be published).

[21] H. Cederquist, H. Andersson, E. Beebe, C. Biedermann, L. Broström, Å. Engström, H. Gao, R. Hutton, J. C. Levin, L. Liljeby, M. Pajek, T. Quinteros, N. Selberg, and P. Sigray, in *Electronic and Atomic Collisions* (Ref. [18]), p. 391.

[22] V. Frohne *et al.* (to be published).

[23] Martin P. Stockli, R. M. Ali, C. L. Cocke, M. L. A. Raphaelian, P. Richard, and T. N. Tipping, Rev. Sci. Instrum. **63**, 2822 (1992).

[24] G. H. Morgan and E. Everhart, Phys. Rev. **128**, 667 (1962); for a review, see B. Fastrup, in *Methods of Experimental Physics: Atomic Physics*, edited by P. Richard (Academic, New York, 1980), Chap. 4.

[25] H. Ryufuku, K. Sasaki, and T. Watanabe, Phys. Rev. A **21**, 745 (1980).

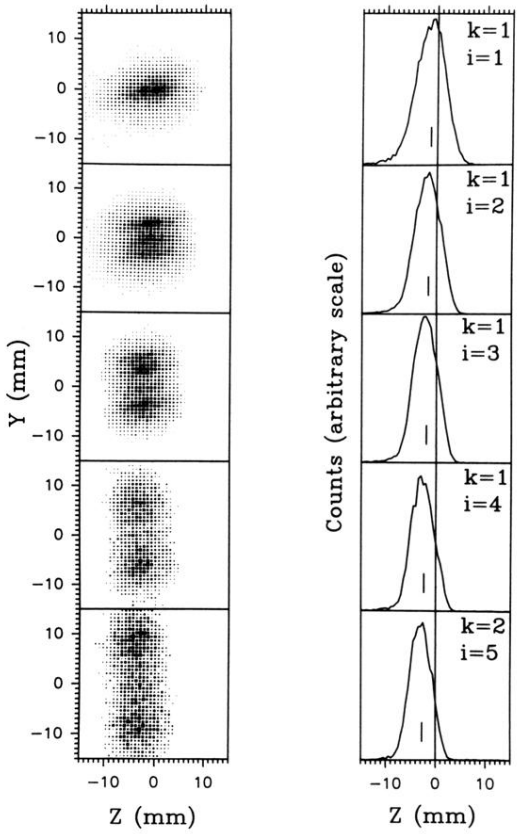
[26] A. Bárány, G. Astner, H. Cederquist, H. Danared, S. Huldt, P. Hvelplund, A. Johnson, H. Knudsen, L. Liljeby, and K.-G. Rensfelt, Nucl. Instrum. Methods Phys. Res., Sect. B **9**, 397 (1985).

[27] A. Niehaus, J. Phys. B **19**, 2925 (1986).

[28] P. Hvelplund, A. Bárány, H. Cederquist, and J. O. K. Pederson, J. Phys. B **20**, 2515 (1987).

[29] H. Cederquist, H. Andersson, G. Astner, P. Hvelplund, and J. O. K. Pedersen, Phys. Rev. Lett. **62**, 1465 (1989).

[30] H. Anderson, H. Cederquist, G. Astner, P. Hvelplund, and J. O. K. Pedersen, Phys. Scr. **42**, 150 (1990).



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