DOUBLE CHARGE EXCHANGE OF ALKALI METAL IONS

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New experimental data on double charge exchange of Li^+, Na^+, and K^+ ions in a number of gases are presented which confirm the possibility of using Massey's adiabatic hypothesis to account for the shapes of the $\sigma_{1-1}(v)$ curves. Conclusions are stated regarding the behavior of some of these curves for $v < v_{max}$ near v_{max}.

INTRODUCTION

In earlier work[1,2] we have shown that double-exchange processes of the type

$$A^+ + B \rightarrow A^- + B^{++} \qquad \text{(I)}$$

obey Massey's adiabatic hypothesis, i.e., the maximum of the $\sigma_{1-1}(v)$ curve is reached at a velocity v_{max} determined from

$$a|\Delta E|/hv_{max} \approx 1, \qquad (1)$$

where a is the range of interaction between the colliding particles,* ΔE is the so-called resonance defect (the change in internal energy of the particles that results from the given process), and h is Planck's constant. a was found to be only slightly dependent on the nature of the colliding particles, and to have the average value 1.5 A.

Similar relations hold true for

$$A + B \rightarrow A^- + B^+ \qquad \text{(II)}$$

(electron capture by fast neutral particles[4-6]), and for

$$A^+ + B \rightarrow A + B^+ \qquad \text{(III)}$$

(ordinary charge exchange[7,8]), with the sole difference that the average values of a for these processes are 3 and 8 A, respectively.

Another important consequence of the adiabatic hypothesis is the theorem that for large values of the adiabatic parameter ($a|\Delta E|hv \gg 1$) the cross sections for inelastic processes are small but increase with velocity according to the formula

$$\sigma = \sigma_0 \exp\{-ka|\Delta E|/hv\}. \qquad (2)$$

Practically nothing is known regarding the shape of the $\sigma(v)$ curve in the adiabatic region $a|\Delta E|/hv \gg 1$. On the basis of data that he had obtained previously[7,8] Hafsted has concluded in reference 9 that $\sigma_{10}(v)$ for ordinary charge exchange obeys Eq. (2) at velocities that are not very far from the maximum.

The present work presents measured cross sections for double charge exchange of Li^+, Na^+, and K^+. These data further confirm (1) for double charge exchange, and also permit some conclusions regarding the shape of $\sigma_{1-1}(v)$ for primary-ion velocities $v < v_{max}$.

EXPERIMENTAL RESULTS

We measured the cross sections σ_{1-1} for $Li^+ \rightarrow Li^-$ in H_2, Ar, Kr, and Xe at 5 — 60 kev, for $Na^+ \rightarrow Na^-$ in H_2, Ar, Kr, and Xe at 10 — 55 kev, and for $K^+ \rightarrow K^-$ in H_2, Ne, Ar, Kr, and Xe at 10 — 80 kev. Beams of alkali metal ions were obtained from either a high-frequency ion source[10] or a thermionic source. The apparatus and technique have been described in references 11 — 14.

Curves of $\sigma_{1-1}(v)$ for $Li^+ \rightarrow Li^-$, $Na^+ \rightarrow Na^-$ and $K^+ \rightarrow K^-$ are shown in Figs. 1 — 3. For each energy the magnitude of σ_{1-1} was obtained by averaging two measurements. For those portions of the curves where low peaks appeared ($Na^+ \rightarrow Na^-$ and $K^+ \rightarrow K^-$ in H_2) and for low velocities, where σ_{1-1} is small, the cross sections were obtained by averaging five or six measurements. The most probable error is ±15%.

A comparison of the $\sigma_{1-1}(v)$ curves for $Li^+ \rightarrow Li^-$ and $Na^+ \rightarrow Na^-$, obtained with the aid of both thermionic and high-frequency sources, shows that the curve shape depends on the type of ion source producing the primary beam. Curves obtained with a high-frequency source exhibit maxima which do not appear on corresponding curves obtained with a thermionic source. As we have shown previously,[1,2] the complex structure of

*A different interpretation of a in (1) has been given by Drukarev.[3]

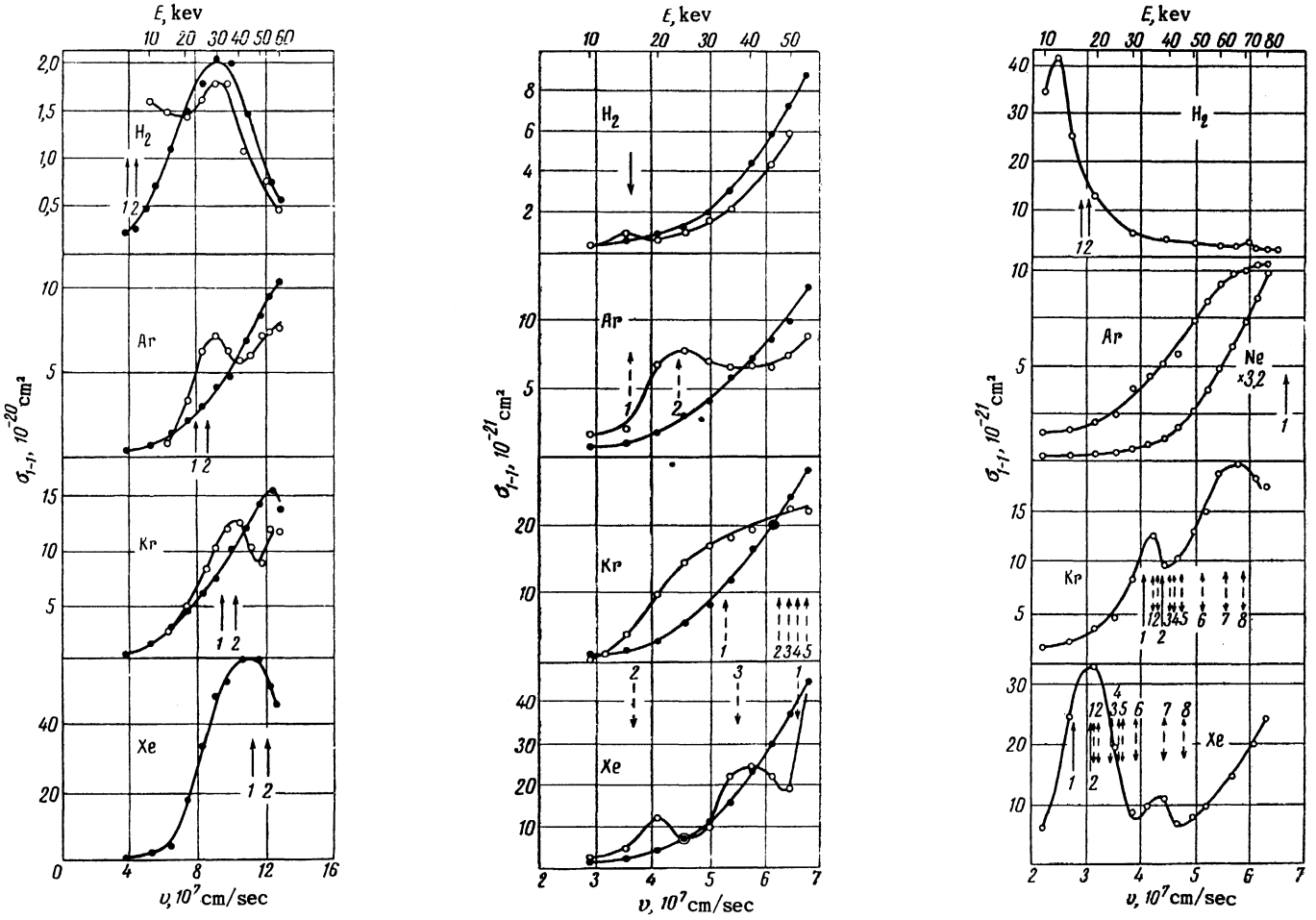

FIG. 1. $Li^+ \to Li^-$ process. ● – thermionic source, o – high-frequency source. Excited levels of fast ions: $1 - 2s^3S_1$; $2 - 2s^1S_0$.

FIG. 2. $Na^+ \to N^-$ process. ● – thermionic source, o – high-frequency source. Excited level of fast ions (process IV in H_2): $3s^5\,^3P^0_{2,0}$. Dashed arrows indicate the following combinations of excited levels of fast and slow ions:

Ar: 1—$Na^+(3s^5\,^3P^0_{2,0}) - Ar^{++}(3p^4\,^1D_2)$, 1— $Na^+(3s^5\,^3P^0_{2,0}) - Kr^{++}(4p^5\,^3P^0_{2,1,0})$,
2—$Na^+(3s^5\,^3P^0_{2,0}) - Ar^{++}(3p^4\,^1S_0)$; 2— $Na^+(3s^5\,^3P^0_{2,0}) - Kr^{++}(4d\,^3D^0_{0,1,2,3,4})$.

Xe: 1—$Na^+(3s^5\,^3P^0_{2,0}) - Xe^{++}(7s\,^5S_2^0)$, Kr: 3—$Na^+(3s^5\,^3P^0_{2,0}) - Kr^{++}(4p^5\,^1P^0_1)$,
2—$Na^+(3s^5\,^3P^0_{2,0}) - Xe^{++}(5d\,^5D_0)$, 4— $Na^+(3s^5\,^3P^0_{2,0}) - Kr^{++}(5s\,^5S^0_2)$,
3—$Na^+(3s^5\,^3P^0_{2,0}) - Xe^{++}(6p\,^5P_3)$; 5— $Na^+(3s^5\,^3P^0_{2,0}) - Kr^{++}(4d\,^3D^0_{3,2,1})$.

FIG. 3. $K^+ \to K^-$ process. ● – thermionic source. Excited levels of fast ions: $1 - 3d^1F^0_3$; $2 - 4s^3P^0_2$. Dashed arrows indicate the following combinations of excited levels of fast and slow ions:

Kr: 1, 2—$K^+(3d\,^1F^0_3) - Kr^{++}(4p^4\,^3P_{1,0})$,
3, 4—$K^+(4s\,^3P^0_2) - Kr^{++}(4p^4\,^3P_{1,0})$,
5—$K^+(3d\,^1F^0_3) - Kr^{++}(4p^4\,^1D_2)$,
6—$K^+(4s\,^3P^0_2) - Kr^{++}(4p^4\,^1D_2)$,
7—$K^+(3d\,^1F^0_3) - Kr^{++}(4p^4\,^1S_0)$,
8—$K^+(4s\,^3P^0_2) - Kr^{++}(4p^4\,^1S_0)$;

Xe: 1, 2— $K^+(3d^1F^0_3) - Xe^{++}(5p^4\,^3P_{2,0})$,
3, 4— $K^+(4s\,^3P^0_2) - Xe^{++}(5p^4\,^3P_{2,1})$,
5—$K^+(3d\,^1F^0_3) - Xe^{++}(5p^4\,^1D_2)$,
6— $K^+(4s\,^3P^0_2) - Xe^{++}(5p^4\,^1D_2)$,
7—$K^+(3d\,^1F^0_3) - Xe^{++}(5p^4\,^1S_0)$,
8—$K^+(4s\,^3P^0_2) - Xe^{++}(5p^4\,^1S_0)$

$\sigma_{1-1}(v)$ curves obtained with high-frequency sources is accounted for by the fact that the primary beam in such cases contains some ions in metastable excited states. During beam passage through a gas, process I (double charge exchange in which only ground-state particles participate) will then be accompanied by other processes:

$$A^{+*} + B \to A^- + B^{++} \qquad \text{(IV)}$$

(double charge exchange of fast excited ions) and

$$A^{+*} + B \to A^- + B^{++*} \qquad \text{(V)}$$

(double charge exchange of fast excited ions with the formation of slow excited ions).

Unlike the cases of $Li^+ \rightarrow Li^-$ and $Na^+ \rightarrow Na^-$, identical $\sigma_{1-1}(v)$ curves for $K^+ \rightarrow K^-$ charge exchange are obtained with high-frequency and thermionic sources, i.e., peaks are observed at identical velocities in the two cases. The difference which is found in the respective heights of these peaks results from different numbers of excited ions in the beams. Since thermionic emission cannot be accompanied by the excitation of K^+ ions evaporating from the emitter (potassium aluminosilicate), excited ions must be produced through collisions of primary ions with residual gas molecules and with the metal surfaces of beam-defining slits and of the magnetic mass-monochromator cover. The apparatus was redesigned for the purpose of determining the part played by residual gas in the excitation of ions in a K^+ beam. The ion beam path from the source to the collision chamber was reduced. The residual gas pressure was considerably reduced by a modified pumping system and by the use of liquid-nitrogen traps. In this manner the number of residual gas molecules in the beam path from the source to the collision chamber was reduced by a factor of about 10, without affecting the $\sigma_{1-1}(v)$ curve for the $K^+ \rightarrow K^-$ process. We can thus conclude that excited ions in a K^+ beam from a thermionic source are produced by collisions with metal surfaces. The absence of excited ions in Li^+ and Na^+ beams from thermionic sources evidently results from the fact that the excitation energy of metastable Li^+ and Na^+ is considerably above that of K^+.

In our earlier work[1,2] we have shown that (1) is applicable to double charge exchange processes and that a is approximately identical, with an average value of 1.5 A, for double charge exchange of unexcited and excited ions. On this basis we have analyzed the $\sigma_{1-1}(v)$ curves for $Na^+ \rightarrow Na^-$ and $K^+ \rightarrow K^-$.

The simplest of the $Na^+ \rightarrow Na$ processes is that of Na^+ in H_2, when excited gas ions cannot result. The peak of the curve obtained with a high-frequency source is associated with an admixture of Na^+ ions in the metastable excited states $3s_5\,^3P_2$ and $3s_3\,^3P_0$ (with very close excitation energies). The shape of $\sigma_{1-1}(v)$ for $Na^+ \rightarrow Na^-$ in Ar, Kr and Xe, using a high-frequency source, can be attributed to double charge exchange of excited Na^+ ions in $3s_5\,^3P_{2,0}$ states, with the simultaneous formation of exeited doubly-charged gas ions. The arrows in Figs. 2 and 3 indicate the positions of the additional peaks associated with processes IV and V (solid arrows for IV and dashed arrows for V). The level of the fast excited ion participating in process IV is stated in the captions of Figs. 1 — 3. The levels of the fast ion and of the gas ion participating in process V are stated in the captions of Figs. 2 and 3.

When $K^+ \rightarrow K^-$ processes were analyzed similarly, the case of K^+ in H_2 was found to be especially interesting. Here the main and secondary peaks associated with charge exchange of K^+ in the metastable states $3d\,^1F_3^0$ and $4s\,^3P_2^0$ are within the investigated velocity range. In Fig. 3 the secondary peak is seen to be considerably higher than the main peak. The explanation for this lies in the rapid falling-off of $\sigma_{1-1\max}$ with increasing resonance defect,[15] while $|\Delta E|$ for $K^+(3p^6\,^1S_0$, the ground state$) \rightarrow K^-$ is larger than for $K^+(3d\,^1F_3^0) \rightarrow K^-$.

The shape of $\sigma_{1-1}(v)$ for $K^+ \rightarrow K^-$ in Kr and Xe can be attributed to double charge exchange of K^+ in $3d\,^1F_3^0$ and $4s\,^3P_2^0$ states, accompanied by the production of excited doubly-charged gas ions. For $K^+(3d\,^1F_3^0) \rightarrow K^-$ in Ar a peak should be found at 90 kev, with all other peaks located at higher energies. Fig. 3 shows a peak at 80 kev for K^+ in Ar. For K^+ in Ne the peak closest to the investigated energy range is located at 360 kev $[K^+(3d\,^1F_3^0) \rightarrow K^-]$; $\sigma_{1-1}(v)$ increases monotonically in the investigated range.

The shapes of $\sigma_{1-1}(v)$ for $Na^+ \rightarrow Na^-$ and $K^+ \rightarrow K^-$, as well as for $H^+ \rightarrow H^-$, $Li^+ \rightarrow Li^-$, $B^+ \rightarrow B^-$, $O^+ \rightarrow O^-$ and $F^+ \rightarrow F^-$, which we had investigated previously, are fully accounted for by the adiabatic criterion (1).

In order to account for the shape of σ_{1-1} at velocities $v < v_{max}$ we may use $\sigma_{1-1}(v)$ for $Li^+ \rightarrow Li^-$ and $Na^+ \rightarrow Na^-$ in the case of a thermionic source, as well as for $K^+ \rightarrow K^-$ in Ne and Ar, since in these cases the velocity region of interest is not distorted by secondary peaks that represent double charge exchange involving excited particles.

It must be remembered that our velocity region $v < v_{max}$ is not adiabatic for any of the aforementioned cases (with the possible exception of K^+ in Ne), since the condition $a|\Delta E|/hv \gg 1$ is not fulfilled. We could therefore not expect $\sigma_{1-1}(v)$ in this region to be described by (2). In actuality, a considerable portion of $\sigma_{1-1}(v)$ almost up to the peak obeys (2) for all of the cases mentioned ($Li^+ \rightarrow Li^-$ and $K^+ \rightarrow K^-$ in Ar). This can be seen from the plot (Fig. 4a) of $\ln \sigma_{1-1}$ as a function of $1/v$ for $Li^+ \rightarrow Li^-$ in H_2; here two points lie off the straight line near the peak of $\sigma_{1-1}(v)$ (which corresponds to the smallest value of $1/v$), and an additional point at the smallest velocity (3.7×10^7 cm/sec). The number of points not lying on the rectilinear segment of $\ln \sigma_{1-1} = f(1/v)$ increases at lower velocities. This is seen for Na^+

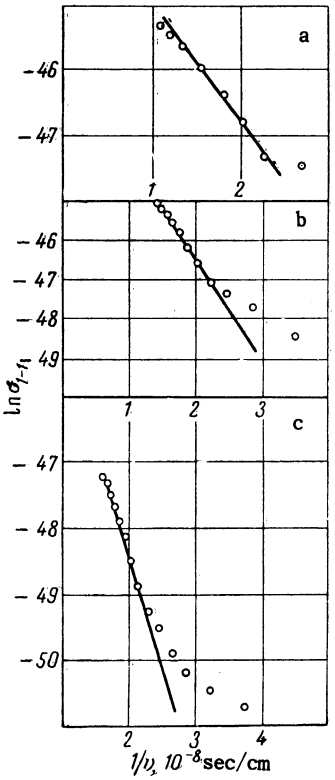

FIG. 4. $\ln \sigma_{1-1} = f(1/v)$ for (a) $Li^+ \rightarrow Li^-$ in H_2; (b) $Na^+ \rightarrow Na^-$ in H_2; (c) $K^+ \rightarrow K^-$ in Ne

$\rightarrow Na^-$ in H_2 (Fig. 4b), where three points lie off the straight line, and is even more prominent for $K^+ \rightarrow K^-$ in Ne (Fig. 4c), where five points lie off the straight line.

The $\sigma_{1-1}(v)$ curve thus obeys (2) wherever the condition $a|\Delta E|/hv \gg 1$ is not satisfied, and ceases to obey (2) wherever the condition is satisfied to any extent ($a|\Delta E|/hv \approx 7$ for the lowest velocity on $\sigma_{1-1}(v)$ in the case of $K^+ \rightarrow K^-$ in Ne). The situations for processes II[15] and III[9] are analogous.

In connection with the observed characteristics of $\sigma_{1-1}(v)$ in the region $v < v_{max}$ for electron capture by fast ions and atoms, during a discussion at the conference on electronic and atomic collisions held in Riga during June of 1959, V. M. Dukel'skiĭ suggested that (2) is not fully obeyed because the relative velocity of particles involved in the given process does not equal their relative velocity before collision. The manner in which points depart from the straight line in plots of $\ln \sigma_{1-1} = f(1/v)$ indicates that if this suggestion is correct, then $v_{true} > v$, where v_{true} is the true velocity at which the process occurs and v is the velocity before collision. The increment $\Delta v = v_{true} - v$ results from the interaction between the colliding particles. The relative velocity change resulting from the acceleration induced by the interaction forces is easily calculated from the departure of points lying off the rectilinear segment of $\ln \sigma_{1-1} = f(1/v)$. From the most extremely deviating point for K^+ in Ne we obtain $\Delta v/v \approx 60\%$, which is too large to result from the known attractive forces between atomic particles. Dukel'skiĭ's suggestion can therefore not account for the way in which $\sigma_{1-1}(v)$ departs from (2).

In disagreement with Hafsted,[9] we maintain that the way in which σ_{1-1} falls off in the region $v < v_{max}$ near the peak according to (2) has no relation to the adiabatic hypothesis, since $a|\Delta E|/hv \gg 1$ is clearly unfulfilled in this velocity region. This condition is evidently not fulfilled even for K^+ in Ne, in which case we obtained the highest values of $a|\Delta E|/hv$. This is evident from the fact that the points of $\ln \sigma_{1-1} = f(1/v)$ that lie off the straight line do not determine a second straight line, which would have been evidence of a true adiabatic region. Moreover, even in a true adiabatic region $\sigma_{1-1}(v)$ might not obey (2) since a can be velocity dependent. The experimental information required for determining $\sigma_{1-1}(v)$ in the adiabatic region is still extremely meager. Therefore the further investigation of $\sigma(v)$ for different processes at low velocities remains a very important task in the physics of atomic collisions.

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