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Search for a possible isotope effect in charge-changing collisions involving hydrogen and deuterium†

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Abstract. Charge equilibrium fractions $F_{1\infty}$, $F_{0\infty}$ and $F_{\bar{1}\infty}$ and relative charge-changing cross sections σ_{10} and σ_{01} were measured for the four possible combinations among hydrogen and deuterium beams and targets, namely H^+–D_2, H^+–H_2, D^+–D_2 and D^+–H_2. Within experimental errors of less than 0·3% for $F_{0\infty}$, 1·2% for $F_{1\infty}$ and 2% for both $F_{\bar{1}\infty}$ and the relative cross sections, no systematic differences for these isotope combinations were found in the velocity range $1{\cdot}38\times10^8$ to $2{\cdot}24\times10^8$ cm s^{-1}. Neglecting the small negative charge fraction, the ratio σ_{10}/σ_{01}, given by $F_{0\infty}/F_{1\infty}$, agrees to within 1·5% for the different combinations studied.

1. Introduction

Theoretical discussions of charge-exchange collisions compared at equal velocities between hydrogen-like projectiles and targets (Jackson and Schiff 1953, Boyd and Dalgarno 1958) do not show a dependence of the corresponding cross sections on the masses of the colliding particles.

However, experiments performed in different laboratories are in disagreement with these theoretical predictions. Fite *et al.* (1958) measured a capture cross section σ_{10} for H^+ in atomic D, larger than for D^+ in D, when compared at equal velocities of the projectiles. Surprisingly, such an isotopic difference was not observed by these authors when the target was molecular.

In view of the results of Fite *et al.*, Hollricher (1965) made a systematic experimental study of the total cross section σ_{10} for the four possible combinations of hydrogen and deuterium beams and molecular targets in search for an isotopic dependence. His results show an enhancement of electron capture for the combination light projectile–heavy target. Apparent isotopic differences between the cases $H^+ + D_2$ and $D^+ + H_2$ amounted up to 10%. Hollricher observed no isotopic difference between the symmetrical cases, H^+ in H_2 and D^+ in D_2. This is in accordance with measurements of Belyaev *et al.* (1967), performed at low ion velocities.

Using beams of ^{3}He and ^{4}He, no isotopic difference in charge equilibrium fractions was observed by Armstrong *et al.* (1965) and Meckbach and Nemirovsky (1967) within error limits of the order of 1%.

The contradictory experimental results for hydrogen and helium isotopes and also the disagreement with theoretical arguments made it desirable to investigate further the possible existence of an isotope effect for hydrogen and deuterium beams and targets.

2. Experimental approach

In order to be able to find eventual small systematic isotopic differences, the search was performed by measuring charge equilibrium fractions of which a direct and precise determination is accessible with relative ease.

According to an argument of Hollricher (1965) the cross section σ_{10} increases, whereas σ_{01} decreases, when the target-to-projectile mass ratio increases. This means that an isotopic effect in the cross sections would be magnified when plotting their ratio. In a

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two-component system this ratio is given by

$$\frac{\sigma_{10}}{\sigma_{01}} = \frac{F_{0\infty}}{F_{1\infty}}.$$

The conditions for a two-component system are approximately fulfilled in the case of a hydrogen or deuterium beam, because the negative component is small. Also, the population of metastable 2S atoms and atoms in highly excited states in the neutral beam were estimated not to exceed 1%.

This work also includes measurements of relative cross sections σ_{10} and σ_{01} which, however, could be compared only for the isotopic combinations with no change of the target gas, that is, H^+–H_2 with D^+–H_2, or H^+–D_2 with D^+–D_2.

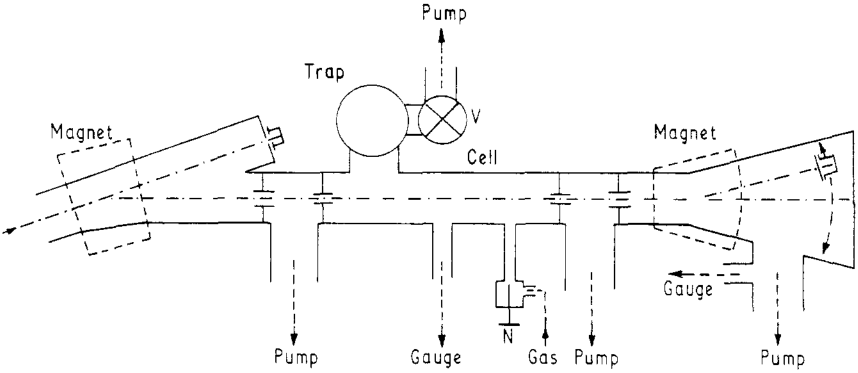

Figure 1. Schematic diagram of experimental equipment.

Figure 1 shows a schematic drawing of the measuring equipment. The H^+ (or D^+) beam, sorted out by magnetic deflection, traverses the differentially pumped interaction cell into which D_2 (or H_2) is injected through a needle valve N. A pumping system with valve V permits rapid outgassing of the cell and, furthermore, by regulating N and V, the establishment of low pressure with a relatively high gas flux. This minimizes gas contamination by internal sources. The purity of the gases was 99·5% or better. After suffering charge changing the beam is magnetically separated into its components which are measured by means of a movable detector. In order to obtain equal detection sensitivity for the beam components of different charge, including the neutrals, each component traversed a thin gold foil mounted in front of the detector, such that newly charge equilibrated beams were measured (Meckbach and Nemirovsky 1967).

The relative cross sections σ_{10} and σ_{01} were determined by measuring the positive fraction $F_1(\pi)$ in a non-equilibrated beam.

According to Hasted (1964) we then have

$$\sigma_{10} = \frac{F_{0\infty}}{\pi} \ln \left\{ \frac{F_{0\infty}}{F_1(\pi) - F_{1\infty}} \right\} = \frac{Q_{10}}{\pi} \quad ; \quad \sigma_{01} = \frac{F_{1\infty}}{\pi} \ln \left\{ \frac{F_{0\infty}}{F_1(\pi) - F_{1\infty}} \right\} = \frac{Q_{01}}{\pi}.$$

π is the target thickness in molecules/cm^2.

To avoid accurate absolute pressure measurements a constant pressure of H_2 was maintained in the interaction cell by fixing the position of the valves N and V (figure 1). By measuring the fraction $F_1(\pi)$ for incident protons and deuterons the value $Q_{10} = \pi\sigma_{10}$ was compared among the isotopic combinations H^+–H_2 and D^+–H_2. As π remained constant an isotopic effect in σ_{10} should appear here. This comparison was also performed for $Q_{01} = \pi\sigma_{01}$.

The same test was performed for the combinations H^+–D_2 and D^+–D_2.

3. Results and discussion

Figure 2 shows the measured charge equilibrium fractions as a function of the ion velocity. The results for the four possible isotopic combinations agree within 0·3% for $F_{0\infty}$, 1·2% for $F_{1\infty}$ and 2% for $F_{\bar{1}\infty}$. They are also in complete coincidence with the values measured by Stier and Barnett (1956) for the case H^+ in H_2.

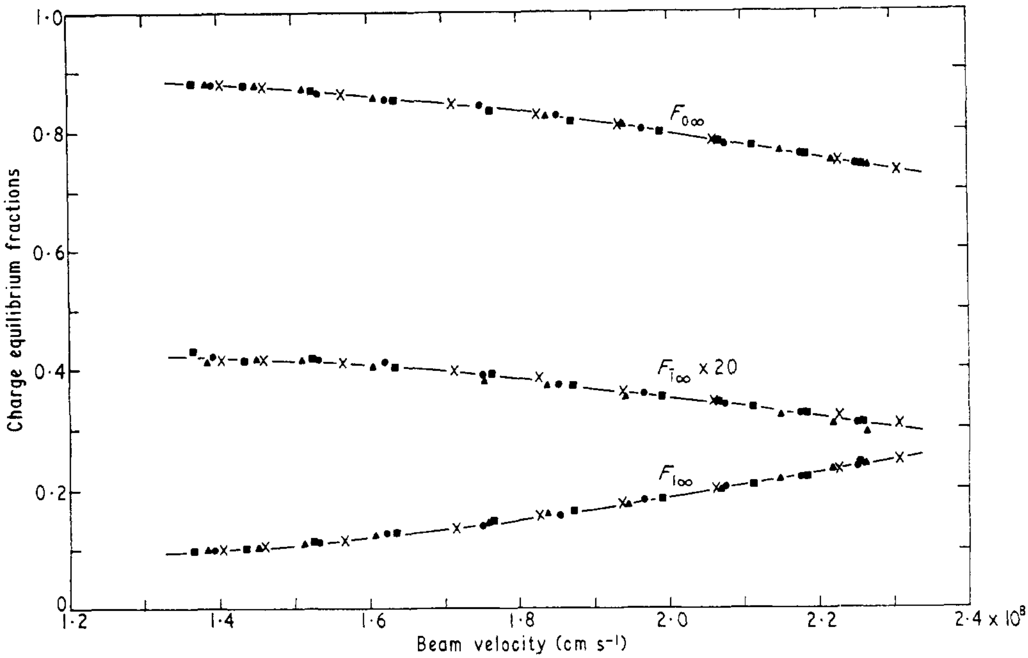

Figure 2. Charge equilibrium fractions for the four possible combinations between hydrogen and deuterium beams and targets. ▲, H^+ in D_2; ×, H^+ in H_2; ■, D^+ in D_2; ●, D^+ in H_2.

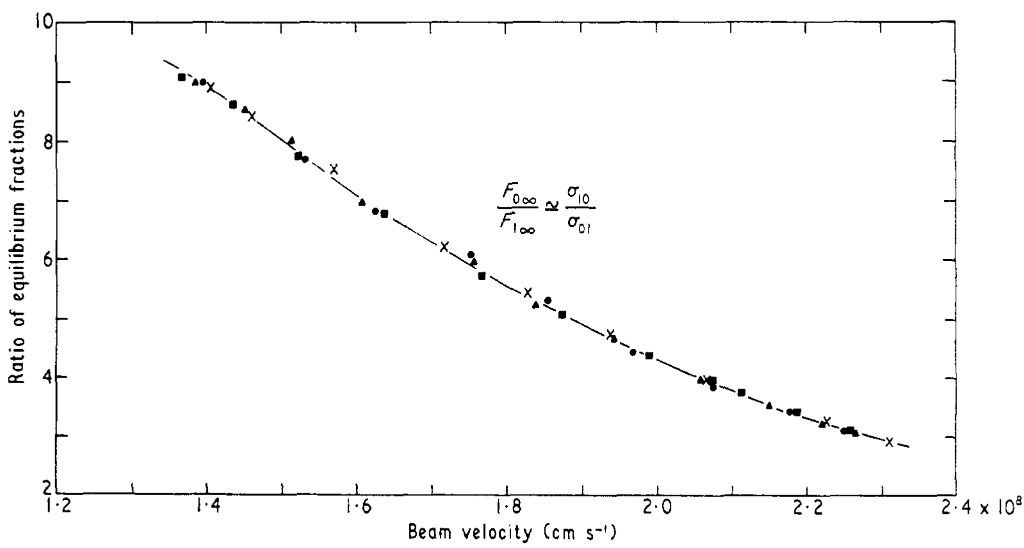

Figure 3. Ratio of charge equilibrium fractions of neutral and monocharged component, approximately equal to the ratio of the cross sections of electron capture and loss, for the four possible isotopic combinations. ▲, H^+ in D_2; ×, H^+ in H_2; ■, D^+ in D_2; ●, D^+ in H_2.

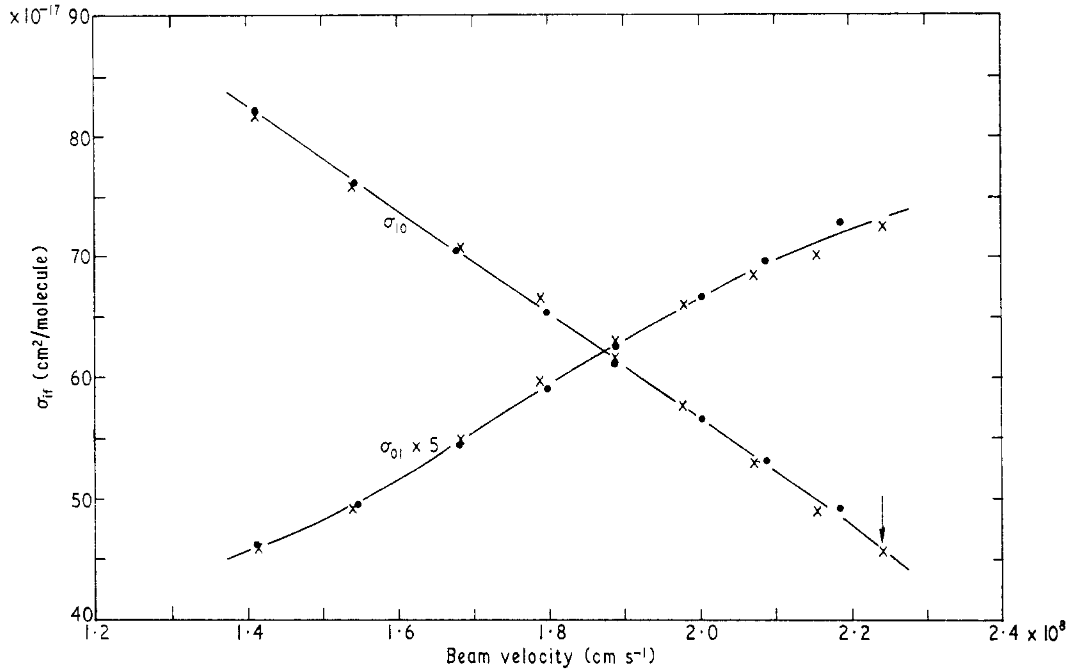

Figure 4. Relative cross sections for electron capture and loss for the combinations H^+ in H_2 and D^+ in H_2. The cross section σ_{10} at $v = 2{\cdot}24 \times 10^8$ cm s^{-1} is normalized to the tabulated value from Allison and García Muñoz (1962). This makes the curve for σ_{10} and also σ_{01} coincide with the tabulated cross sections throughout the velocity range covered. ×, H^+ in H_2; ●, D^+ in H_2.

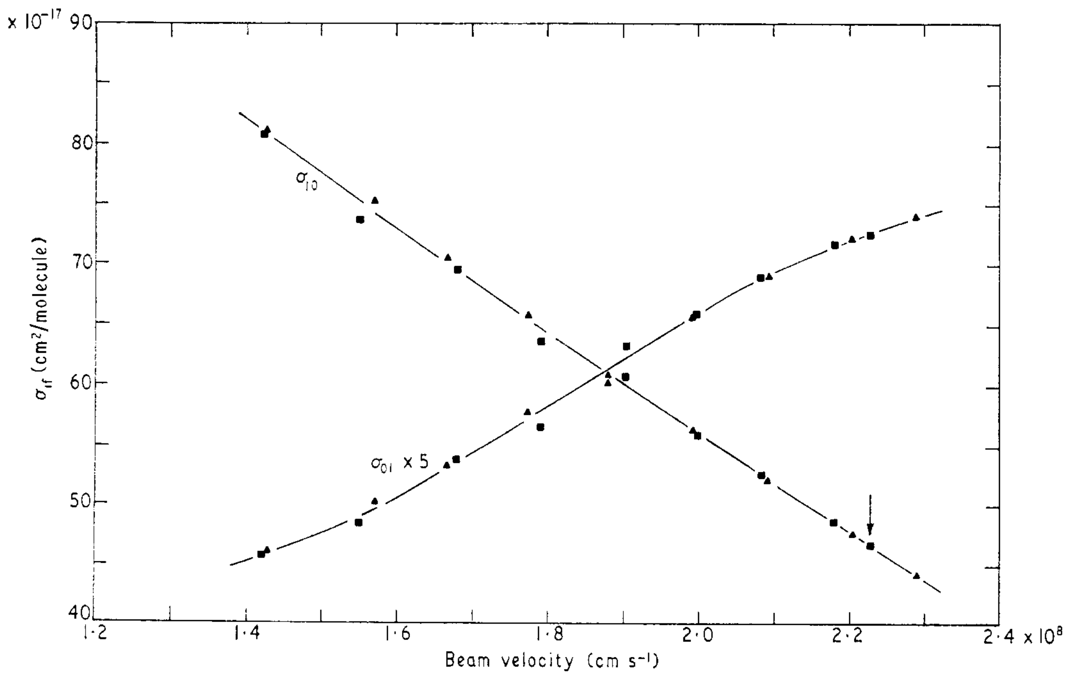

Figure 5. Relative cross sections for electron capture and loss for the combinations H^+ in D_2 and D^+ in D_2. The normalization has been performed as in the preceding case at $v = 2{\cdot}23 \times 10^8$ cm s^{-1}. ▲, H^+ in D_2; ■, D^+ in D_2.

Figure 3 shows the ratio $F_{0\infty}/F_{1\infty} \simeq \sigma_{10}/\sigma_{01}$ as a function of velocity for the four isotopic combinations measured. Within the error limits of 1·5% no systematic difference can be observed.

Relative cross sections σ_{10} and σ_{01} are plotted in figures 4 and 5 after normalizing the σ_{10} value for a given velocity. The coincidence with the values tabulated by Allison and García Muñoz (1962) is within our error limits throughout the velocity range covered. No systematic deviations in the cross sections obtained with hydrogen and deuterium beams are observed within the experimental error of 2%.

The present measurements show that, within the error limits quoted, there is no isotopic dependence in the charge equilibrium fractions and total cross sections in charge-changing collisions involving hydrogen and deuterium beams and molecular targets.

This, having been shown for molecular targets, must hold also in the physically more simple case of an atomic target gas, since even if, as shown by Tuan and Gerjuoy (1960), the Bragg rule does not hold, the isotopic differences in molecular eigenstates could only enhance the possible existence of an isotope effect.

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