

Normalization of Cross Sections for Charge-Transfer Collisions Producing Metastable (2S) Hydrogen Atoms*

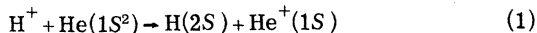
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Reported measurements of the cross section for the production of metastable hydrogen atoms in the 2S state are compared to theoretical predictions for charge-transfer collisions between protons and helium and atomic hydrogen. The magnitudes of the cross sections for some of the experimental data are arbitrarily adjusted to produce a consistent energy dependence of the reaction cross sections. Agreement between recent theoretical predictions and experiment is then found to be excellent, leading to the conclusion that for these excited-state charge-transfer reactions, the Born approximation is not a valid description, even at projectile energies above 100 keV.

The cross section for the production of metastable hydrogen atoms in the 2S state according to the reaction



has been experimentally measured and reported by five independent investigators.¹⁻⁵ These measurements cover a range of projectile impact energies from 3 to 200 keV. The reaction cross section has also been theoretically evaluated twice, a Born approximation calculation⁶ and a recent coupled-channel calculation.⁷ To evaluate the extent of agreement between the various experimental results and the theoretical calculations, a survey of the experimental results has been made. The five experimental measurements are briefly summarized below with the results shown in Fig. 1.

A. Colli *et al.*¹ (7.2 to 39.5 keV) used a Thoneman ion source with no mass separation of the projectile ions. Lyman-alpha radiation from the electric field quenched metastable atoms was detected by a vacuum magnetic photomultiplier for the far *uv*. The cross section was normalized to the Born approximation calculation at 39.5 keV. Displayed errors are assumed to be statistical and systematic only.

B. Jaecks *et al.*² (3 to 23 keV) used a mass analyzed projectile ion beam produced by an electron bombardment ion source. Lyman-alpha radiation from the electric field quenched metastable atoms was detected by an oxygen filtered iodine vapor and helium filled photon counter with a lithium fluoride window. The cross section was measured relative to the 2P state capture cross section for the same reaction which was in turn normalized to the reaction $e^- + \text{H}_2 \rightarrow$ (countable ultraviolet), whose magnitude was based on measurements of the reaction $e^- + \text{H} \rightarrow$ (Lyman alpha). Displayed errors reflect an uncertainty in normalization ($\pm 45\%$) and a statistical or reproducibility error ($\pm 5\%$).

C. Andreev *et al.*³ (10 to 40 keV): Lyman-alpha radiation from the electric field quenched metastable atoms was isolated by a vacuum monochromator and detected by a sodium salicylate coated photomultiplier tube. The cross section was normalized to that for the 2P state capture process in the H^+ -Ne reaction which in turn was absolutely determined by measuring the Lyman-alpha radiation flux with a nitric oxide filled ionization chamber. Displayed errors include absolute measurement uncertainty ($\pm 20\%$) and a statistical error ($\pm 5\%$).

D. Dose⁴ (3 to 70 keV): Lyman-alpha radiation from the electric field quenched metastable atoms was detected by an unfiltered iodine vapor and helium filled photon counter with a lithium fluoride window. The cross section was measured relative to the 2P state capture cross section for the same

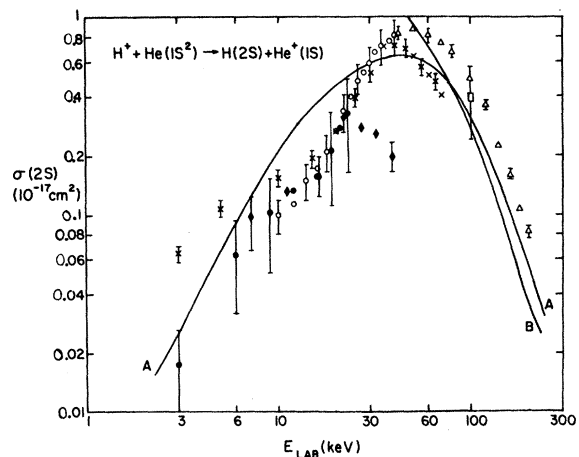


FIG. 1. 2S capture cross sections for protons in helium. Experimental results: \blacklozenge Colli *et al.*, Ref. 1; \bullet Jaecks *et al.*, Ref. 2; \circ Andreev *et al.*, Ref. 3; \times Dose, Ref. 4; \triangle Ryding *et al.*, Ref. 5. Theoretical results: curve A, Sin Fai Lam, Ref. 7; curve B, Born approximation, Mapleton, Ref. 6.

reaction which was in turn normalized to the Born approximation calculation at 70 keV. Displayed errors are assumed to reflect statistical and systematic errors only.

E. Ryding *et al.*⁵ (42.5 to 200 keV): Lyman-alpha radiation from the electric field quenched metastable atoms was detected by an unfiltered silver-magnesium surface secondary emission multiplier detector with a lithium fluoride window. Displayed errors are assumed to reflect systematic and statistical errors only.

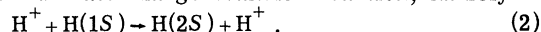
From the above it is seen that only two of the experimental investigations (Jaecks *et al.* and Andreev *et al.*) utilized a method of normalization of the reaction cross section independent of a Born-approximation calculation for either the 2S or 2P state capture reaction. The agreement between these two investigations in both shape and magnitude of the cross section and the lack of agreement among the remaining three investigators then suggests that the Born approximation is not a correct basis for normalization. The experimental data presented in Fig. 1 have been normalized as follows: data of Ryding normalized to the data of Andreev at 42.5 keV (using an extension of the curve of Andreev); data of Dose normalized to Andreev at 20 keV; data of Colli normalized to the average of Andreev-Jaecks at 22 keV. Included in Fig. 1 are the coupled channel calculation and the Born-approximation calculation (curves A and B, respectively). With the exception of the high-energy data of Colli and the low- and high-energy data of Dose, a consistent picture emerges for the energy behavior of the cross section for Reaction (1).

Without knowing the details of the experimental procedures used by Dose, it is not possible to ascribe the differences between his data and the other investigators to experimental error. However in the case of Colli *et al.*, it appears that failure to ensure a pure mass species in the projectile ion beam is a probable cause of the observed discrepancy between the data of these investigators and that of Andreev and Jaecks.

From Fig. 1 it is seen that the coupled channel calculation, rather than the Born-approximation calculation best describes this collision process. While not all observed features of the cross section are reproduced by the calculation (in particular the "shoulder" at 10 keV) the position of the cross-section maximum and its over-all magnitude are in reasonable agreement with the observations. Even at the highest projectile energies reported here the Born approximation fails to agree with observations and seriously underestimates the observed cross section. We therefore conclude that for the rearrangement collision discussed here the Born approximation is not valid, a conclusion not unexpected on theoretical grounds.

The conclusion presented above prompted the investigation of the published data for another

excited-state charge-transfer reaction, namely



This reaction has been experimentally investigated only twice^{5,9} but has been theoretically evaluated many times using a number of approximation techniques.

We again briefly describe the experimental techniques used to observe this reaction.

A. Bayfield⁹ (3 to 22.5 keV): Lyman-alpha radiation from the electric field quenched metastable atoms was detected by an unfiltered Bendix magnetic electron multiplier with a tungsten photocathode. The target hydrogen atoms were formed in a heated tubular furnace through which the ion beam passed. The dissociation fraction, that is the ratio of hydrogen atoms to hydrogen molecules, was quoted as 0.97 ± 0.03 . The cross section was normalized to the 2S state capture cross section for the H^+ -Ar reaction using the data of Jaecks and Andreev. Displayed errors include all systematic and statistical errors but do not include an estimated uncertainty of $\pm 15\%$ in the H^+ -Ar reaction cross section.

B. Ryding *et al.*⁵ (42.5 to 120 keV): Detection techniques are identical to those used for the H^+ -He reaction by the same investigators. The source of target hydrogen atoms was a tubular furnace similar to that used by Bayfield. The dissociation fraction was not specified. The cross section was normalized to the Born-approximation calculation at 100 keV. Displayed errors are assumed to reflect systematic and statistical errors only.

Figure 2 displays the results of Bayfield, as

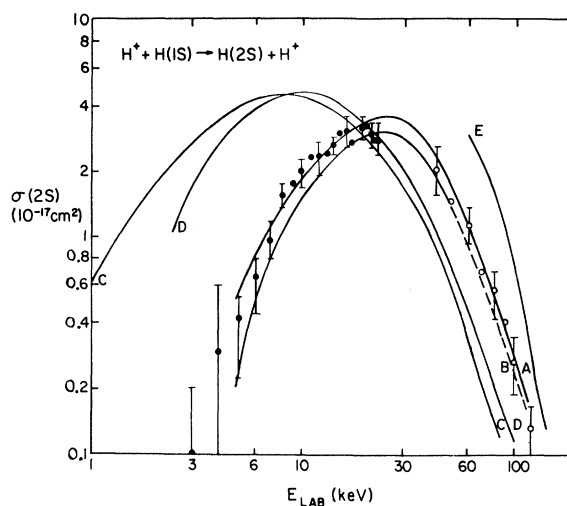


FIG. 2. 2S capture cross sections for protons in atomic hydrogen. Experimental results: \circ Ryding *et al.*, Ref. 5; \bullet Bayfield, Ref. 9. Theoretical results: curve A, Willets and Gallaher, Ref. 10; curve B, Lovell and McElroy [solid part 1SA/2SA/2SB (3 state), dashed part 1SA/2SB (2 state)], Ref. 15; curve C, Coleman and Trelease, Ref. 13; curve D, Bates and Dalgarno, Ref. 11; curve E, Mapleton, Ref. 12.

published, and those of Ryding, normalized to the H^+ -He reaction data. The agreement between the two sets of experimental data and curve A, the 4 state coupled-channel calculation (hydrogenic representation) of Wilets and Gallaher¹⁰ is at once obvious and striking. The high-energy behavior of Bayfield's data is assumed to be scatter only and not a tendency toward agreement with curve D (Born approximation^{11,12}).

As in the H^+ -He reaction, the Born approximation seriously underestimates the cross section, even at the highest energy and displays serious disagreement with experiment respecting magnitude and position of the cross-section maximum. This behavior is consistent with theoretical arguments regarding the convergence of the Born expansion for rearrangement collisions.¹³ A recent impulse approximation calculation¹⁴ is included in Fig. 2 as curve C, and also fails to predict the observed features of the cross section. This impulse approximation calculation was originally thought to exhibit good agreement with experiment for the $2P$ state capture cross section for the H^+ -H reaction, especially at low energies (1 to 10 keV), but recent measurements¹⁵ of this

reaction tend to negate those conclusions. Included for reference in Fig. 2 are the close coupling calculations of Lovell and McElroy,¹⁶ curve B, using 2 and 3 states in the hydrogenic representation. The Brinkman-Kramers approximation¹² is also included as curve E, chiefly for historic reasons.

As in the case of Reaction (1) we observe, in the light of the data in Fig. 2, that the Born approximation is not a valid description of the type of rearrangement collision described by reaction (2). It is tempting at this point to claim the existence of sufficient experimental evidence to invalidate the Born approximation for all rearrangement collisions, however recent observations of total charge-transfer collisions between protons and helium and certain other target gases¹⁷ show excellent agreement with the Born approximation at energies of 100 to 2500 keV. Thus we must limit our statement to include only excited-state charge-capture collisions.

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