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## Comments

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# Excitation transfer in ion–Rydberg-atom collisions

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Recently, electron-loss cross sections were presented by Kim and Meyer [Phys. Rev. Lett. **44**, 1047 (1980)] for 40 keV/amu $N^{3+} + H^{**}(n)$ collisions which scaled as $n^{3.12}$, where $n$ is the principal quantum number of the excited $H^0$. Such results are in contrast to an $n^2$ scaling predicted by classical and first Born theoretical methods. Our calculations indicate that a major component of the experimentally observed ion signal was due to Stark ionization by deflector grids of highly excited $H^0$ produced in excitation-transfer collisions. Inclusion of the excitation process in a theoretical interpretation reveals qualitative agreement between theory and experiment and stresses the importance of excitation transfer in ion–Rydberg-atom collisions.

Recently, Kim and Meyer[1] reported measurements of the cross sections for electron removal from hydrogenic Rydberg atoms by collisions with $N^{3+}$:

$$N^{3+} + H^{**}(n) \rightarrow \begin{cases} N^{3+} + H^+ + e^- & \text{(1a)} \\ N^{2+} + H^+ . & \text{(1b)} \end{cases}$$

Reaction (1a) is impact ionization while (1b) is the electron-capture process. The experiment was conducted at a velocity of $2.8\times 10^8$ cm/sec for principal quantum numbers $n$ ranging from 9 to 24. For these collisions, the ratio of heavy-particle-collision velocity to that of the Rydberg orbital electron $v/v_e$ varies from 11.5 to 30.7. Thus, it is accurate to predict that the electron-removal process will be dominated by impact ionization, reaction (1a).

Analysis of the experimental results by Kim and Meyer indicates the cross section for the sum of (1a) and (1b) varies as

$$\sigma = 1.19\times 10^{-15}\, n^{3.12}\ \text{cm}^2 \tag{2}$$

for $9 \leqslant n \leqslant 24$. This result is in contrast to classical-trajectory Monte Carlo (CTMC) calculations[2] which predict the electron-removal cross section for $n \geqslant 4$ should vary as

$$\sigma = 2.9\times 10^{-15} n^2\ \text{cm}^2 . \tag{3}$$

Thus, there is a large difference between theoretical predictions and experimental measurements in the $n$-scaling dependence and in the magnitude of the cross sections. The experimental values lie a factor of 5 to 15 above the theoretical results given by Eq. (3) for $n = 9$ to 24. A slight amount of the difference can be compensated by the fact that the CTMC results, because of neglect of quantum tunneling, underestimate first Born calculations[3] by ~ 30% in the $v/v_e$ range of 10 to 30. However, a major discrepancy remains.

We suggest the source of the difference may be due to ion–Rydberg atom-excitation-transfer collisions. Excitation cross sections are extremely large and scale with the geometric size of the Rydberg atom, $n$.[4] Furthermore, high-lying Rydberg atoms produced in the collision region can be readily ionized by subsequent electric fields.

To determine the possible effect of excitation-transfer reactions, it is necessary to examine the details of the measurement. Experimentally, it is necessary to deflect the $H^+$ formed via Reactions (1a) and (1b) into an analyzer for counting. However, the deflector voltages used in the $N^{3+} + H^{**}$ measurements, 1–1.5 kV/cm, are such that they will also Stark ionize excited $H^{**}$ formed in excitation-transfer collisions

$$N^{3+} + H^{**}(n) \rightarrow N^{3+} + H^{**}(n') , \tag{4}$$

with final $n'$ values greater than 25 (1.5 kV/cm) or 28 (1 kV/cm). Thus, the true cross section measured by Kim and Meyer was the sum of electron loss and excitation to $n' \gtrsim 27$.

In order to determine the relative importance of the excitation process, we have calculated the 40 keV/amu $N^{3+} + H^{**}$ electron-loss and excitation-transfer cross sections pertinent to the experimental results:

$$\sigma = \sigma_{\text{loss}} + \sigma_{\text{excit}}(n \rightarrow n' \geqslant 27) . \tag{5}$$

The electron-loss cross sections were taken from Ref. 2 and the excitation-transfer cross sections

 

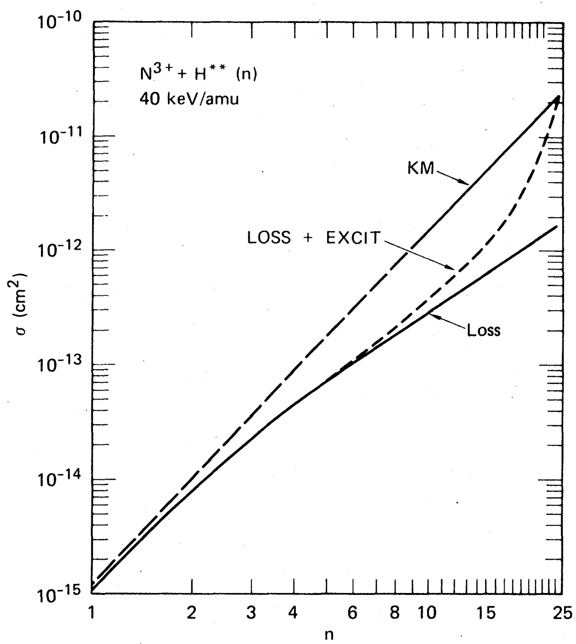


FIG. 1. Cross sections for collisions of $N^3 + H^{**}$ ($n$) at 40 keV/amu. The values obtained from the analysis of experimental data by Kim and Meyer (Ref. 1) are denoted by KM. The long dashed line is an extrapolation of the cross-section dependence given in Ref. 1 for $n \leq 8$. The theoretical CTMC calculations for the electron-loss process, reactions (1), are given by the line denoted LOSS while the electron-loss plus excitation-transfer cross section defined by Eq. (5) is labeled LOSS + EXCIT.

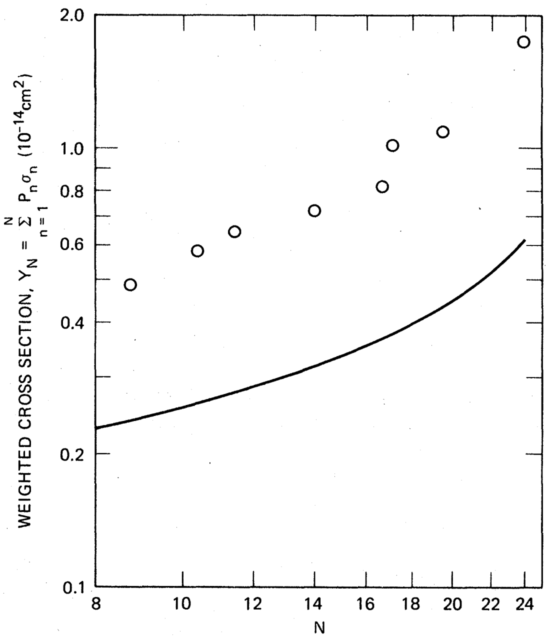


FIG. 2. Population-weighted cross section as a function of $N$, the upper cutoff of $n$. The circles are the data of Kim and Meyer (Ref. 1) while the line was calculated using the LOSS + EXCIT cross section given in Fig. 1 combined with $P_n = 0.41/n^3$.

were obtained for the $N^{3+} + H^{**}$ system by performing additional CTMC calculations. The exact classical method is known[4] to give very accurate values of excitation cross sections for large changes in $n$. Moreover, when $\Delta n/n$ is small the analytical formulas given by Lodge *et al.*[4] are accurate and compare well to CTMC results.[2] The results of the CTMC calculations are compared to the experimentally derived cross sections in Fig. 1. When the theoretical cross sections are related to the experimental setup of Kim and Meyer via the use of Eq. (5), we find the excitation cross sections are 25% larger than the electron-loss values at $n = 9$, with the magnitude of the excitation component rising to equal that of the electron loss at $n = 16$ and an order-of-magnitude larger at $n = 24$.

Since the agreement between the theoretical and experimental cross sections is poor, it is necessary to directly compare to the experimentally observed quantity, the population-weighted cross section

$$Y_N = \sum_{n=1}^{N} P_n \sigma_n , \quad (6)$$

where $N$ is the upper cutoff of the field ionizer. We have used the same population distribution for the $H^{**}$ beam as was observed by Kim and Meyer in the $9 \leq n \leq 24$ region,

$$P_n = 0.41/n^3 . \quad (7)$$

The use of the functional dependence given by Eq. (7) for values of $n \leq 8$ to obtain the cross-section dependence of Eq. (2) is quite questionable in that the sum of all $P_n$ is only 0.50 rather than the desired 1.0 value. However, in absence of a more accurate distribution function, we use Eq. (7) with the anticipation that our calculated $Y_N$ values will necessarily lie below the experimental results.

The comparison between the theoretical and experimental observed population-weighted cross sections is shown in Fig. 2. The agreement is approximately a factor of 2 with similar slopes for both theoretical and experimental $Y_N$. We should emphasize that the theoretical calculations were made using the cross section defined by Eq. (5) for the sum of electron loss and excitation transfer. We attribute the major reason why the theoretical values underestimate the experimental values as due to the use of Eq. (7) which underestimates the contribution from the low $n$ states by up to a factor of 2.

Thus, there appears to be no serious discrep-

ancy between theory and experiment as stated in Ref. 1. More importantly, however, these calculations and the $N^{3+} + H^{**}$ and other[5] experimental data stress the need to consider excitation processes in the design of an experimental apparatus for ion–Rydberg-atom ionization measurements. Even within the safeguard of varying the deflector voltages, changes in the experimental cross sections may not be observed due to the large contribution of the excitation cross section for production of very highly excited Rydberg levels.

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