Interferences in electron emission spectra from 1, 3 and 5 MeV $H^+$ + $N_2$ collisions

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Abstract. Electron interferences associated with coherent two-centre emission in 1-5 MeV $H^+$ + $N_2$ collisions are investigated. Spectra were measured for ejected electrons with energies from 5-410 eV and observation angles in the range 30\textdegree to 150\textdegree. Experimental molecular $N_2$ cross sections were normalized to theoretical atomic $N$ cross sections, revealing oscillatory structures that do not change significantly with observation angle or collision velocity. It is suggested that the oscillations are due to previously observed secondary interferences arising from intramolecular scattering following ejection of a K-shell electron.

1. Introduction

Previously, we investigated electron interferences for fast $H^+$ ($v/c \sim 0.1$) and $Kr^{34+}$ ($v/c \sim 0.3$) ions interacting with $H_2$, revealing both primary Young-type interferences due to coherent electron emission from the identical atomic centers as well as secondary interferences caused by intramolecular scattering \cite{1,2,3,4}. The primary oscillations were found to depend strongly on the electron observation angle \cite{2,4} and to a lesser extent on the collision velocity \cite{4}, in general agreement with theoretical calculations \cite{5,6,7}. In contrast, the secondary oscillations which have two to three times higher oscillation frequencies showed little variation with either the emission angle or the collision velocity \cite{3,4}. These results have prompted new studies for $H_2$ by other investigators \cite{8,9,10,11}.

Electron interferences for diatomic molecules other than $H_2$ have been studied for ejection of valence electrons from $N_2$ and $O_2$ \cite{12} and core (K-shell) electrons from $N_2$ by photons \cite{13}. In the latter case, the interference structures were attributed to intramolecular scattering following K-shell ejection. In the present work, our previous studies for $H^+ + H_2$ \cite{4} are extended to $N_2$ for which electron ejection is expected to occur primarily from the L-shell with a binding energy nearly identical to that of $H_2$, but ionization can also occur from the K-shell. Since the internuclear separation of $N_2$ is larger than that of $H_2$ (2.1 a.u. compared to 1.4 a.u.), equivalent to increasing the slit separation in Young’s experiment, higher oscillation frequencies for the primary interference structures are expected for $N_2$. Notably, for photons \cite{12,13} the incident energy can be chosen to selectively ionize the K or L shell, whereas fast ions can ionize either shell for a given collision energy.

Here, electron emission spectra for 1, 3 and 5 MeV $H^+$ + $N_2$ collisions were measured for ejected electron energies ranging from 5-410 eV and observation angles 30\textdegree to 150\textdegree. The measured $N_2$ cross sections divided by the corresponding theoretical atomic $N$ cross sections show oscillatory structures that exhibit little dependence on the observation angle or collision velocity, a result that suggests the structures are due to secondary oscillations arising from intramolecular scattering.
2. Theory

To reveal the expected small amplitude interference structures superimposed on exponentially decreasing cross sections, the measured molecular \( \text{N}_2 \) cross sections are divided by corresponding theoretical atomic \( \text{N} \) cross sections, i.e., \[1\]

\[
\left( \frac{\sigma_{\text{N}_2}}{\sigma_{\text{N}}} \right)_{\text{norm}} = \frac{d^2 \sigma_{\text{N}_2}}{d\Omega d\epsilon} / \frac{d^2 \sigma_{\text{N}}}{d\Omega d\epsilon}
\]

where \( d\Omega \) is the solid angle and \( d\epsilon \) refers to the outgoing electron energy. The cross section \( \sigma_{\text{N}_2} \) denotes the molecular two-center emission and \( \sigma_{\text{N}} \) is the cross section for independent emission from the two \( \text{N} \) atoms. Theoretical cross sections for atomic \( \text{N} \) were calculated in this work and normalized to the measured \( \text{N}_2 \) cross sections according to equation (1).

To compare the experimental ratios obtained from equation (1) with the corresponding theoretical ratios, electron emission cross sections for \( \text{N}_2 \) must be calculated including the expected effects due to interference. Preliminary calculations were made using the previous model for \( \text{H}_2 \) \[5\]. However, the situation is much more complex for \( \text{N}_2 \). Specifically, there are six molecular orbitals from which the electron may be ejected with different symmetries and ionization potentials. The positions of the interference maxima and minima depend on the ionization potentials, causing the expected experimental interference ratio to be a superposition of several curves with different frequencies. Nevertheless, these preliminary results indicate primary oscillation frequencies that vary with angle in a manner similar to \( \text{H}_2 \) \[5\].

Based on the formulation of Ref. \[5\], the normalized ratios of the interference structures calculated from equation (1) can be parameterized according to the damped sinusoidal function

\[
f(k) = A \left[ 1 + \sin \left( kcd \right) / kcd \right] + B
\]

where \( k \) is the ejected electron velocity (momentum), \( d \) is the molecular internuclear separation, and \( c \) is a fitting parameter representing the frequency. The constants \( A \) and \( B \) \((A + B \approx 1)\) represent the interfering and non-interfering contributions to the normalized cross section ratio, respectively. It has been shown that the normalized ratios are well represented by this functional form \[2,3,4\].

3. Experimental procedure

Measurements were conducted at Western Michigan University using the 6-MV tandem Van de Graaff accelerator. \( \text{H}^+ \) beams with energies 1, 3 and 5 MeV were collimated to a diameter of about 2 mm and directed into the scattering chamber onto an \( \text{N}_2 \) target with a diameter of ~2-3 mm. Collimated beam intensities ranged from ~40-500 nA. The \( \text{N}_2 \) target was supplied by a gas jet located ~3.5 mm above the center of the beam line and the flow rate was adjusted to maintain an average pressure of \( 5 \times 10^{-5} \) Torr in the scattering chamber to ensure single collision conditions. The background pressure in the chamber was \( 1 \times 10^{-6} \) Torr. Electrons emitted from the target were measured for ejection energies \( 5 - 410 \) eV and observation angles 30°, 45°, 60°, 90°, 120°, 135° and 150° with respect to the incident beam direction using a parallel-plate electron spectrometer equipped with a channel electron multiplier (CEM). Stray magnetic fields inside the scattering chamber were minimized by shielding with a \( \mu \)-metal liner. The electron spectra were normalized to the incident beam intensity. Background spectra were recorded with no target gas to correct for residual gas events.

4. Results and discussion

The measured \( \text{N}_2 \) electron emission yields were normalized to theoretical atomic \( \text{N} \) cross sections according to equation (1), as shown in figure 1 for 3 MeV \( \text{H}^+ + \text{N}_2 \) at three observation angles. The shapes of the calculated cross sections generally agree with the measured yields except at higher electron energies where the theory falls off faster than the measured data, and also for low electron energies at 60°. Some of the high-energy discrepancy may be due to the contribution of the K-auger peak near 350 eV and also the low energy tail of the well-known binary encounter peak that results from direct encounters between the projectile and a target electron.
Ratios of the experimental molecular $\text{N}_2$ cross sections to the theoretical atomic N cross sections from figure 1 are shown in figure 2. The ratios show structures with evidence of oscillatory behavior. The rising trend of the ratios at high electron energies is due to the noted discrepancy between the data and the theory in figure 1, and has been observed previously for $\text{Kr}^{34+} + \text{H}_2$ collisions [1]. Further analysis is needed in the regions of higher electron velocity to eliminate the discrepancies with the atomic N theory. Similar results were obtained for the other angles investigated.

Figure 1. Experimental $\text{N}_2$ cross sections normalized to theoretical atomic N cross sections for 3 MeV $\text{H}^+ + \text{N}_2$ at the indicated electron observation angles. The pronounced peaks are due to auger transitions occurring at $\sim 351$ eV and $\sim 371$ eV, respectively.

Figure 2. Measured molecular $\text{N}_2$ cross sections divided by theoretical atomic N cross sections for 1, 3 and 5 MeV $\text{H}^+$ ions at the indicated observation angles. The smooth curves represent the results of fitting the damped sinusoidal function given by equation (2) to the ratios. The resulting frequency parameter $c$ is shown in each case. The estimated uncertainties in the frequency parameter are about $\pm 0.1$.

The smooth curves in figure 2 show the results of fitting equation (2) to the cross section ratios. The striking feature of the fitting results is that the values obtained for the frequency parameter $c$ show no significant dependence on the electron observation angle or the collision velocity, as shown in figure 3. Furthermore, the oscillation frequencies for $\text{N}_2$ ($\Delta k \sim 2$ a.u.) as determined from the $c$ values are significantly higher than expected for the primary interferences in $\text{N}_2$ based on the molecular internuclear separation compared to $\text{H}_2$, and are in fact nearly identical with the secondary frequencies previously found for $\text{H}^+ + \text{H}_2$ collisions [4].

Figure 3. Frequency parameter $c$ from equation (2) for 1, 3 and 5 MeV projectile energies and electron observation angles 30°, 45°, 60°, 90°, 120°, 135° and 150° for $\text{H}^+ + \text{N}_2$. The values of $c$ for the three energies were averaged because they showed no significant variation with energy. The uncertainties in $c$ are about $\pm 0.1$. 
These results suggest that the observed structures may be due to secondary oscillations resulting from intramolecular scattering. It will be recalled that Rolles et al [13] observed only secondary interference effects for K-shell photoemission from N\textsubscript{2}. If the present structures are indeed due to secondary interferences, then it must be questioned why no primary Young-type interferences are seen. Perhaps N\textsubscript{2} ionization involving a valence (L-shell) electron doesn't lead to primary interferences because this electron doesn't ‘see’ two distinct atomic N centers when it is ejected due to the large screening by the remaining 13 electrons. Hence, there can be no coherent emission for these valence electrons. On the other hand, the core (K-shell) electrons are bound to one center or the other and consequently there can be no primary interference for these electrons, only secondary interference when the primary ionization ‘wave’ from one atomic center interferes with this same wave subsequently scattered at the other center. In support of this hypothesis, preliminary calculations based on the formalism presented in [5] show that the interference term has opposite signs for bound and anti-bound molecular orbitals. If these orbitals are close in energy (as for the K-shell), the interference terms from the two orbitals cancel out, and we obtain independent ejections from the two centers. However, such an explanation for the present case of H\textsuperscript{+} + N\textsubscript{2} collisions in which both K- and L-shell electrons are ejected needs further theoretical and experimental investigation.

5. Conclusion

Measured N\textsubscript{2} cross sections for electron emission normalized to corresponding theoretical atomic N cross sections for fast H\textsuperscript{+} + N\textsubscript{2} collisions reveal oscillatory behavior that exhibits little dependence on the electron observation angle or the collision velocity. By fitting the normalized ratios to a damped sinusoidal function, oscillation frequencies characteristic of secondary interferences previously attributed to intramolecular scattering are obtained, while no evidence for primary Young-type interferences is found. It is suggested that the observed structures are due to ejection of a K-shell electron in which direct emission interferes with emission following scattering at the other atomic center. Further studies are needed to substantiate these ideas.

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