Ionizing Collisions of Protons with Ammonia & Methane

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Semi-empirical relations have been employed to obtain differential and total ionization cross-sections due to proton impact on ammonia and methane. Peaks appearing in the experimental cross-sections have been explained as due to the Auger process. The results so obtained are in fair agreement with the available experimental data.

1. Introduction

The study of ionization of ammonia \((\text{NH}_3)\) and methane \((\text{CH}_4)\) is of importance to interpret the various phenomena occurring in a number of planetary atmospheres, particularly that of Jupiter. When a proton of sufficient energy is absorbed in a gas, it loses a large fraction of its energy in the ionizing collisions and electrons are produced. These ejected electrons produce additional ionizations and excitations. Hence, to study the energy loss of proton in gases, a knowledge of energy loss cross-section or the ionization cross-section per unit energy range is required.

In principle, the ionization problem can be investigated quantum mechanically. However, even with the present fast electronic computers, it is a formidable task due to mathematical complexities. For most of the atomic targets, the ionization calculations are limited to first Born approximation. For molecules the situation is worse. Ionization of \(\text{CH}_4\) by electron impact has been investigated by Inokuti\(^{1}\) considering the first Born approximation. However, the results are only in qualitative agreement with the experimental data. In general, the theory overestimates the cross-sections. So far no theoretical investigation of differential cross-sections for ionization of \(\text{NH}_3\) and \(\text{CH}_4\) due to proton impact is done. Recently, Lynch et al.\(^{2}\) have carried out experimental investigations for such ionizations. Hence, in the absence of quantum mechanical investigations, which are quite involved, some simplified approach yielding reasonable results for the above mentioned processes will be of interest. Recently, Khare and Kumar\(^{3}\) have employed semi-empirical approach to investigate differential ionization cross-section and total ionization cross-section for nitrogen gas due to proton impact. The results obtained are in good agreement with the available experimental data. In the present work, we have employed the same approach to study the ionization process of \(\text{NH}_3\) and \(\text{CH}_4\) due to proton impact. To our knowledge this is the first theoretical investigation for the ionization of \(\text{NH}_3\) and \(\text{CH}_4\) due to proton impact.

2. Theory

According to Born approximation, at high energy, ionization cross-section \(\sigma_\text{I} (E)\) due to proton impact is equal to the corresponding cross-section, \(\sigma_\text{e} (E)\), due to incident electron having same velocity,\(^{4,6}\) i.e.

\[
\sigma_\text{I} (E) = \sigma_\text{e} (E) \quad \text{with} \quad E_\text{e} = \frac{m}{M} E \quad \text{...(1)}
\]

where \(E\) and \(E_\text{e}\) are the energies of proton and electron, respectively, the corresponding masses being \(M\) and \(m\).

Using the semi-empirical approach of Khare and Kumar\(^{3}\) we have, for the ionization cross-section per unit energy range \(Q_\text{I} (E,\epsilon)\) or loss cross-section, \(S_\text{I} (E, W_\text{e})\), due to proton impact

\[
Q_\text{I} (E,\epsilon) = S_\text{I} (E, W_\text{e}) = f_{\text{I}1} (E,\epsilon) S_{\text{RI}} (E, W_\text{e})
+ f_{\text{I}2} (E,\epsilon) S_{\text{RP}} (E, W_\text{e}) \quad \text{...(2)}
\]

with
\[ f_{p1} (E, \varepsilon) = \frac{1}{1 + \frac{M}{m} \frac{W_p}{E}} e^{-\frac{W_p}{E}} \ln \left[ 1 + \frac{m}{M} \frac{W_p}{E} \right] \]

\[ f_{p2} (E, \varepsilon) = \frac{e^\varepsilon}{e^\varepsilon + e_{ep}^2} \]

The Born-Bethe cross-section \( S_{BB} (E, W_p) \) and Rutherford loss cross-section \( S_{RP} (E, W_p) \) are, respectively, given by

\[ S_{BB} (E, W_p) = \frac{4n^2 \alpha^2 R^2 M}{m E W_p} \frac{\partial f (W_p, 0)}{\partial W_p} \ln \left[ \frac{m}{M} CE \right] \]

\[ S_{RP} (E, W_p) = \frac{4n^2 \alpha^2 R^2 M}{m E} \left[ \frac{S}{W_p - I} \right] \]

where \( \alpha \) is the first Bohr radius, \( R \) the Rydberg constant, \( S \) the number of ionizable electrons in the target excluding \( K \) shell electrons, \( W_p \) the transfer of energy in the collision and \( e_{ep} \) is a parameter chosen in such a way that there is a good agreement between the theoretical results and the experimental data over a wide range of energy. For the present investigation, a suitable value for \( e_{ep} \) is equal to 60 eV. This value is consistent with the switch over of \( e_0 \) taken by Khare. It is assumed that the energy loss \( W_p \) is equal to \( \varepsilon + I \) where \( \varepsilon \) is the energy of the ejected electron and \( I \) is the ionization potential of the target. The quantity \( \frac{\partial f (W_p, 0)}{\partial W_p} \) is the optical oscillator strength for the absorption of a photon of energy \( W_p \). To simplify the calculations for \( \text{NH}_3 \) we have fitted the experimental data of Van der Wiel and Brion for the optical oscillator strength with the following simple relations

\[ \frac{\partial f (W_p, 0)}{\partial W_p} = m W_p + K \]

for \( I \leq W_p < 16 \text{ eV} \) with \( m = 0.0285 \text{ eV}^{-1} \),

\[ K = -0.177 \text{ eV}^{-1} \]

and

\[ \frac{\partial f (W_p, 0)}{\partial W_p} = A \exp (-B W_p) \]

with

\[ A = 0.6992 \text{ eV}^{-1}, B = 0.052 \text{ eV}^{-1} \]

for \( 16 \leq W_p < 60 \text{ eV} \)

where

\[ A = 0.1755 \text{ eV}^{-1}, B = 0.02618 \text{ eV}^{-1} \]

for \( W_p \geq 60 \text{ eV} \)

Similarly, the experimental data for optical oscillator strength of \( \text{CH}_4 \) given by Backx et al. are fitted with the following simple relations.

\[ \frac{\partial f (W_p, 0)}{\partial W_p} = m W_p + K \]

for \( I \leq W_p \leq 13.4 \text{ eV} \)

with

\[ m = 0.0933 \text{ eV}^{-1}, K = -0.79 \text{ eV}^{-1} \]

and

\[ \frac{\partial f (W_p, 0)}{\partial W_p} = A \exp (-B W_p) \]

with

\[ A = 1.2586 \text{ eV}^{-1}, B = 0.07719 \text{ eV}^{-1} \]

for \( 13.4 < W_p < 40 \text{ eV} \); and

\[ A = 0.2528 \text{ eV}^{-1}, B = 0.357 \text{ eV}^{-1} \]

for \( W_p > 40 \text{ eV} \)

In Eqs. (3) and (5) it is assumed that the collisional parameter \( C \) is independent of \( W_p \). The values of the parameters \( I, C \) and \( S \) employed in the present investigations are shown in Table 1. For electrons having energy \( E \geq 1 \text{ keV} \), the relativistic corrections have been applied.

The above energy spectrum has been integrated from 0 to \( W_{\text{max}} \) to obtain the total ionization cross-section \( Q_i (E) \). The following expression for \( W_{\text{max}} \) given by McGuire and Edgar et al. is used.

\[ W_{\text{max}} = 4 \frac{m}{M} E \]

Hence, we have

\[ Q_i (E) = \int_0^{W_{\text{max}}} Q_p (E, \varepsilon) d\varepsilon \]

Substituting the value of \( Q_p (E, \varepsilon) \) of Eq. (2) in Eq. (8), we have obtained \( Q_i (E) \).

In Eq. (2) the contribution of the \( K \)-shell is not included. However, \( K \)-shell ionization gives rise to the production of Auger electrons. For \( \text{NH}_3 \),

<table>
<thead>
<tr>
<th>Target</th>
<th>( I )</th>
<th>( C )</th>
<th>( S )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{NH}_3 )</td>
<td>10.15</td>
<td>0.066 (Ref. 11)</td>
<td>8</td>
</tr>
<tr>
<td>( \text{CH}_4 )</td>
<td>13.00</td>
<td>0.094 (Ref. 12)</td>
<td>8</td>
</tr>
</tbody>
</table>
the Auger electrons of energy about 365 eV are emitted from nitrogen atom. Similarly, the energy of the Auger electrons produced from CH₄ is about 250 eV due to carbon atom.² The ionization cross-section per unit energy range due to Auger electrons, \( Q_A(E, \varepsilon) \), is given by the following relation as employed by Khare and Kumar³

\[
Q_A(E, \varepsilon) = Q(E, \bar{\varepsilon}) \exp\left(-\frac{|\varepsilon - \bar{\varepsilon}|}{\Delta}\right) \quad \text{(9)}
\]

where \( \varepsilon \) is 365 and 250 eV for NH₃ and CH₄, respectively. An appropriate value of \( \Delta \) seems to be 25 eV. The term \( Q(E, \varepsilon) \) is the difference between the peak value of differential cross-section of Lynch et al.² and our present value obtained from Eq.(2) at \( \varepsilon = 365 \) and 250 eV for NH₃ and CH₄, respectively. These differential ionization cross-sections due to Auger electrons are added to the differential ionization cross-section calculated by using Eq.(2).

To investigate the contribution of Auger electrons to the total ionization cross-section, \( Q_A(E, \varepsilon) \) in Eq. (9) is integrated over \( \varepsilon \) from zero to infinity. Thus, the total K-shell ionization cross-section \( Q_K(E) \) is given as follows.

\[
Q_K(E) = \Delta \left(2 - e^{-\bar{\varepsilon}/\Delta}\right) Q(E, \varepsilon) \quad \text{(10)}
\]

The results so obtained from Eq. (10) are added to the total ionization cross-section calculated by using Eq. (8).

3. Results and Discussion

In the present investigation, we have calculated the values of \( Q_p(E, \varepsilon) \) using Eq. (2) for proton impact ionization of ammonia and methane for proton energies 0.25, 1.0, and 2.0 MeV. These are shown in Figs. 1-4 by solid curves. It is evident from

Fig. 1—Variation of differential ionization cross-section with the ejected electron energy \( \varepsilon \) for NH₃

Fig. 2—Same as Fig. 1 but for CH₄

Fig. 3—Variation of differential ionization cross-section with the ejected electron energy \( \varepsilon \) for CH₄

Fig. 4—Same as Fig. 3 but for Auger electron contributions also (The actual values may be obtained by multiplying by a factor 10 at energy \( E=2\text{MeV} \))
ejection from nitrogen atom in NH₃ and carbon atom in CH₄. We have taken this into account in Eq. (9). Our theoretical results including the contribution of Auger electrons to differential cross-sections shown by dash-dot curves, are in good agreement (within 20%) with the experimental data of Lynch et al.² shown by crosses.

Using Eqs. (8) and (10) we have calculated total ionization cross-sections for NH₃ and CH₄ with the incident proton energy varying from ionization threshold to 10 MeV. These are shown in Figs. 5 and 6. These values are compared with the experimental values of Lynch et al.² available at three proton impact energies equal to 0.25, 1.0 and 2.0 MeV shown by crosses. Our present values are thus in good agreement with these experimental data (within 12% for NH₃ and 30% for CH₄). It may be noted that the uncertainty in the experimental measurement of \( Q_p (E, \epsilon) \) and \( Q_{1p} (E) \) is about 20%.

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References