Theoretical calculation of total cross sections for $e^+ - \text{NH}_3$ molecule at low energies

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Abstract. A parameter-free spherical complex optical potential model is used to calculate the differential and total cross sections for the scattering of positrons by the NH$_3$ molecules in the energy range 10 eV - 400 eV. The optical potential is constructed from a near-Hartree-Fock one-center expansion of ammonia (NH$_3$) wave function and it is then treated exactly in a partial-wave analysis involving variable phase approach to extract complex phase shifts. The present theoretically calculated results show good agreement with the available experimental results for total cross sections at positron energy $\geq$ 30 eV.

1. Introduction

The total cross sections (including elastic plus energetically possible all inelastic channels) for electron/positron collisions with a large number of atoms and polyatomic molecules have been experimentally measured from as low as (~ 1 eV) to keV energy region [1-3]. Theoretical description of positron scattering from molecules is given by Armour [4]. These cross sections have important applications in space, plasma, laser, biomedical science and the environment. In particular, the discovery of NH$_3$ molecules in the interstellar medium has increased the attention paid to its spectroscopic and dynamical properties [5], whereas in the field of plasma chemistry, NH$_3$ has gained importance as it is a source of nitrogen atoms for the fabrication of nitride films and the synthesis of other nitrogen compounds [6]. On the theoretical side, to study the scattering of electrons and positrons with diatomic and polyatomic molecules, many approaches ranging from as simple as independent atom model (IAM) [7] to ab initio methods [8] have been proposed and developed. In case of positrons colliding with ammonia molecules, IAM approach was used by Raizada and Baluja [9]. In their calculations, total cross section for positron impact on a molecule was expressed as an incoherent sum over the total cross sections of the atoms of the molecules. The rule is called as the additivity rule. It has also been applied for the electron impact on the various molecules by many workers [10-12]. On the experimental side, total cross sections for positrons and electrons colliding with NH$_3$ molecules were measured by Sueoka et al [13] using time-of-flight (TOF) method in the energy range 1 – 400 eV. They had applied a magnetic field of 4.5 G in the range of 1 – 7.5 eV, of 9 G in the range of 0.7 – 400 eV in their experimental technique. The experimental cross sections were obtained by a normalization method. As Ab initio calculations are quite difficult to perform in case of polyatomic molecules, therefore, here our goal is to apply a spherical complex optical potential (SCOP) approach for calculating differential cross sections (DCS) and total ($\sigma$) for positrons colliding with NH$_3$ molecules in the energy range 10 – 400 eV.
2. Methodology

The total interaction of the positron molecule system can be represented by a local complex optical potential, namely,

\[ V_{\text{opt}}(r) = V_R(r) + i V_{\text{Abs}}(r) \]  

where the real part \( V_R(r) \) of the optical model potential, consists of the static potential \( V_{\text{st}} \) and parameter free correlation polarization potential \( V_{\text{cop}} \). The complex optical potential is obtained by adding a model absorption potential \( V_{\text{abs}}(r) \) as imaginary part to the real potential. All the potentials can be generated easily once the target charge density \( \rho_0(r) \) is known.

The ammonia molecule belongs to the \( C_3v \) symmetry point group (electronic \( ^1A_1 \) ground state) with the electronic configuration \( 1a_1^22a_1^21e^43a_1^2 \). A single-center expansion technique (obtained from Jain and Thompson [14]) with a nitrogen atom at the center is employed for all orbitals in the near-Hartree-Fock limit using experimental values for nuclear geometry (such as bond length 1.012Å and bond angle = 106.7º). We determine \( \rho_0(r) \) with enough terms in the expansion of each orbital. \( \rho_0(r) \) is expanded in terms of symmetry-adapted functions belonging to the totally symmetric \(^1A_1\) irreducible representation of the molecular \( C_3v \) point group

\[ \rho_0(r) = \sum \tilde{\rho}_{LH}(r) X_{LH}^{A_1}(\hat{r}) \]  

In the spherical approximation, we only need the first term \((L = 0, H = 1)\) of the expansion of equation (2), in order to evaluate all the three terms, i.e. \( V_{\text{st}}, V_{\text{cop}} \) and \( V_{\text{abs}} \). Explicit expression for \( V_{\text{st}} \) is given in the literature (see, for example, Gianturco and Jain [15]).

The attractive term i.e. polarization effect, which arises due to distortion in the target charge cloud because of incoming charge particle. This effect is invoked by a parameter free correlation polarization potential, which is based on the correlation energy of the target molecule. The calculation of an actual polarization potential which provides the second and higher order effects in the positron - molecule interaction is rather a difficult task. Following Padial and Norcross [16] and Gianturco et al [17] the correlation polarization potential, for various range of radial distance \( r \) is expressed in terms of the density parameter \( r_s \)

\[ r_s(r) = \left[ \frac{3}{4\pi \rho_0(r)} \right]^{1/3} \]

as,

\[ V_{\text{cop}}(r) = \begin{cases} 0.0311 \ln r_s + 0.006 r_s \ln r_s - 0.015 r_s - 0.0584, & 0.7 \leq r_s \leq 10.0 \\ 0.02224 \ln r_s - 0.07356, & 10.0 \leq r_s \leq 1.0 \end{cases} \]

The imaginary part of the optical model potential is referred as absorption potential, which takes into account the flux going through all possible inelastic channels. An exact determination of \( V_{\text{abs}} \) is a very difficult task. Thus a number of empirical and semi-empirical models have been proposed. In the present work, it is determined \textit{ab initio} as the function of charge density of the target molecule following the procedure of Staszewska et al [18]. This is written as

\[ V_{\text{abs}}(r) = -\rho_0(r)\left(\nu_{\text{loc}}/2\right)^{1/2} \left(8\pi/10k_f^3\right)XH(k^2 - k_f^2 + 2\Delta)(A_1 + A_2 + A_3) \]

Where,

\[ \nu_{\text{loc}} = \frac{1}{2} k^2 - V_{\text{st}} - V_{\text{pol}} \]

\[ A_1 = 5k_f^3/2\Delta \]

\[ A_2 = -k_f^2(5k_f^2 - 3k_f^2)/(k^2 - k_f^2)^2 \]

\[ A_3 = 2H(2k_f^2 + 2\Delta - k_f^2)X[(2k_f^2 + 2\Delta - k_f^2)^{5/2}/(k^2 - k_f^2)^2] \]
Here $H(x)$ is a Heaviside function defined by

$$H(x) = \begin{cases} 1, & x \geq 0 \\ 0, & x < 0 \end{cases}$$

where $\Delta$ is the mean excitation energy which is obtained in the closure approximation from the ground-state target wave function $\psi_0$ of the target molecule.

$$\Delta = \frac{2 \langle \psi_0 | z^2 | \psi_0 \rangle}{a_o}$$

For NH$_3$, $\Delta$ is calculated to be 14.986 eV.

Finally, a partial wave analysis approach is adopted to compute the $\ell^0$ partial wave phase shifts for the solution of radial Schrodinger equation and a variable-phase approach (VPA) [19] and has been employed to find its solution. The corresponding quantities (i.e. DCS and $\sigma_t$) are then easily obtained from S-matrix at each energy. All our cross sections are converged with respect to the number of partial waves up to a value of 0.001 radians only.

3. Results and discussion

In figure 1, DCS curves have been shown at incident energy 50 eV. At this energy, neither theoretical calculation nor experimental measurements for positrons scattering with ammonia molecule is available in the literature. Therefore, we have calculated DCS by using both complex and real potential in order to study the influence of the absorption potential. Noticeably, in the present fixed-nuclei approximation the DCS’s at $\theta = 0^\circ$ are not defined. It is clear from the figure that the absence of absorption potential reduces the DCS in magnitude, which is characterized by a dip in the angular distribution at the angles 50$^\circ$ and 65$^\circ$.

![Figure 1.](image1.png)  
**Figure 1.** Differential cross sections for $e^+ - \text{NH}_3$ scattering. Present calculations: _____, with absorption; -----, without absorption.

![Figure 2.](image2.png)  
**Figure 2.** Total cross-sections ($\sigma_t$) for $e^+ - \text{NH}_3$ scattering. Calculations: _____, present results; ----, Raizada and Baluja [9]. Experiment: X, Sueoka *et al* [13].

Total cross sections are also evaluated from the DCS’s. Further, one can avoid the forward angle-peaking behavior of the DCS’s by calculating the corresponding momentum-transfer cross section due to the $(1 - \cos \theta)$ term. In figure 2, the $\sigma_t$ cross sections for $e^+ - \text{NH}_3$ are presented and compared with the IAM calculations of Raizada and Baluja [9] together with experimental measurements of Sueoka *et al* [13]. It is observed that our theoretical results are in better agreement with the experimental data at all energies except below 30 eV. At lower energies, the disagreement may be due to the contribution of the dipole, quadrupole etc. terms, which plays an important role in non-spherical targets. The
comparison between our results and the IAM calculations [9] points out that the effect of bonding is dominant factor in this energy region.

4. Conclusions
It is finally concluded that a parameter-free model potential constructed for the full interaction of the collision system within the framework of the spherical-complex-optical model potential approach is quite adequate to yield the differential and total cross sections. The calculated total cross sections are in good agreement with the available experimental measurements.

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