

01 Jul 2015

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Recommended Citation

Z. N. Ozer et al., "Interference Effects for Intermediate Energy Electron-Impact Ionization of H₂ and N₂ Molecules," *Journal of Physics: Conference Series*, vol. 601, no. 1, Institute of Physics - IOP Publishing, Jul 2015.

The definitive version is available at <https://doi.org/10.1088/1742-6596/601/1/012003>

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Interference effects for intermediate energy electron-impact ionization of H₂ and N₂ molecules

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Abstract. We have studied electron impact ionization of H₂ and N₂ molecules at intermediate energies to look for possible two center interference effects experimentally and theoretically. Here we report a study of the interference factor I for 250 eV electron-impact ionization. The experimental measurements are performed using a crossed-beam-type electron-electron coincidence spectrometer and theoretical calculations are obtained using the Molecular Three Body Distorted Wave Approximation (M3DW). We found that the I-factor demonstrated strong evidence for two-center interference effects for both H₂ and N₂. We also found that the I-factor is more sensitive to projectile angular scans than to ejected electron energy scans which indicate that for the present set of kinematics the diffraction of the projectile from two scattering centers is more important than interference between electron waves emitted from two different centers.

1. Introduction

Electron impact ionization cross sections have been measured and calculated since the early days of collision physics due to the wealth of information that can be obtained about the collision dynamics and also due to their relevance in many application areas. Because of the importance of the cross sections in biological applications, much emphasis has been devoted recently to the experimental studies of electron-impact ionization cross sections of molecules and radicals. The study of the energy and angular distributions of electrons ejected by electron impact is a sensitive means of testing the theory of collision processes.

The possibility of a diatomic molecule acting as a microscopic double slit for photon impact was suggested by Cohen and Fano several decades ago [1]. Since quantum particles are also waves, the next natural question was whether Young's type interference effects could also be observable in triple differential cross section (TDCS) spectra for electron-impact ionization of diatomic molecules. Young type interference effects resulting from the coherent superposition of the scattered waves from two atomic centers were predicted by Stia et al. [2-3] and by Gao et al. [4] for electron impact ionization of molecular H₂ and molecular N₂, respectively. However, due to the small cross sections, the experimental investigations of the ionization of small molecules by this technique are limited. There have been two experimental studies, in coplanar asymmetric geometry, presented for electron impact ionization of H₂ (Milne-Brownlie et al. in 2006 [5] and Casagrande et al. in 2008 [6]). These studies found evidence for interference effects by comparing the relative sizes of the binary and recoil peaks.



The theoretical basis for these studies lies in the interference factor which is defined to be the ratio of the molecular cross section divided by the cross section for two atoms. The idea is that dividing by the atomic cross section should isolate the molecular two-center effects. Cohen and Fano [1] showed that, for photon impact on H₂, this factor could be approximated as

$$I^{CF} = 1 + \frac{\sin(\chi \rho_0)}{\chi \rho_0}$$

where ρ_0 denotes the equilibrium internuclear vector of the molecular target and χ is the momentum transfer. Stia et al. [2-3] showed that the same approximation could be used for electron impact ionization of H₂. This factor predicts that the recoil peaks for atomic H should be either larger or smaller than molecular H₂ depending on the kinematics. Due to the difficulty in measuring atomic H cross sections, the experimentalists substituted He for two atomic H and found the predicted enhancement/suppression in the recoil peak.

Gao et al. [4] predicted similar interference effects for ionization of the $3\sigma_g$ state of N₂. Murray et al. [7] and Hargreaves et al. [8] performed experiments for ionization of N₂ for both asymmetric and symmetric geometries. However, their results did not provide strong evidence for these interference effects.

There are three different types of two center interference effects for electron-impact scattering: (1) Incident electron being diffracted by two scattering centers; (2) scattered electron wave being emitted from two centers; and (3) ejected electron wave being emitted from two centers. We performed calculations for the three different types of possible interference effects for ionization of H₂ and we found that the most important contribution comes from the incident projectile diffracting from two scattering centers [9-10]. As an extension of this study, we have now examined electron impact ionization of N₂ molecules at intermediate energies to look for interference effects both experimentally and theoretically. The experimental measurements are performed using a crossed-beam-type electron-electron coincidence spectrometer and theoretical calculations are obtained using the Molecular Three Body Distorted Wave Approximation (M3DW) [11].

2. Experimental Apparatus

The measurements were performed at e-COL laboratory (Afyon, Turkey) using an electron spectrometer that is designed for electron-electron coincidence (e,2e) experiments. The details of the electron spectrometer are described in detail in previous papers [9, 12-14]. Briefly, the electron spectrometer consists of a electron gun producing a beam of electrons which passes through the gas target perpendicularly, two hemispherical electrostatic energy analyzers, a Faraday cup and a data acquisition system (see in figure 1a). The spectrometer is contained in a cylindrical stainless steel vacuum chamber and the pressure in the chamber is around $\sim 2 \times 10^{-6}$ mbar while the experiment is running. The spectrometer operated at an electron current of $\sim 2 \mu\text{A}$ with a resolution of ~ 0.7 eV. The (e,2e) technique is used to detect two outgoing electrons in coincidence after the ionization of the target. Two electrons of the desired energies are detected and amplified using Channel Electron Multipliers (CEM). This technique has an advantage of obtaining single ionization events meaning the outgoing electrons have originated from the same ionization event. To do this, time correlation between the detected electrons are taken into consideration and time delay between the electrons is converted to a signal that is measured by computer, and a narrow coincidence peak in the timing spectrum is observed. Coincidence electronics are shown in figure 1b.

The overall energy resolution of the coincidence system was limited by both the thermal spread of the electrons emitted from the tungsten hairpin cathode and the analyzer system. By measuring the full width at half-maximum (FWHM) of the binding energy spectra, the coincidence energy resolution was found to be ≈ 1.5 eV. This resolution is good enough to separate the $3\sigma_g$ orbital of the N₂ molecule.

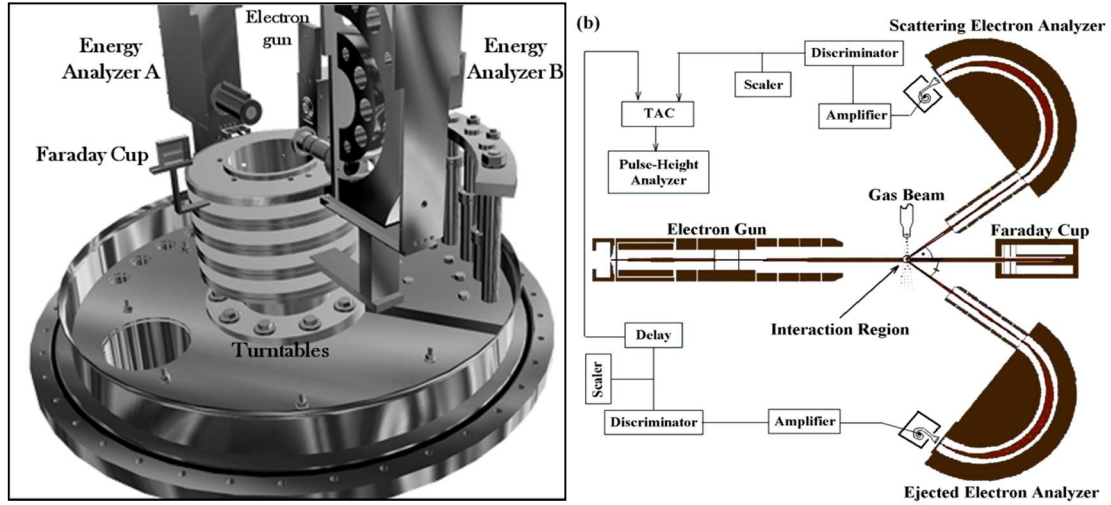


Figure 1. Sketch of electron spectrometer. The main components are electron gun, two electron analyzers and Faraday cup that are mounted on the turntables (a). Coincidence electronics used to accumulating a coincidence timing spectrum at each kinematics (b).

3. Theory

Al-Hagan et al. [11] showed that the molecular three-body distorted wave approximation (M3DW) coupled with an orientation-averaged molecular orbital approximation [15] yielded a good agreement with experimental TDCS data for H₂. The molecular 3-body distorted wave (M3DW) approximation has been presented elsewhere [16-17] so only a brief description of the theory will be presented. The triple differential cross section (TDCS) for the M3DW is given by:

$$\frac{d\sigma}{d\Omega_a d\Omega_b dE_b} = \frac{1}{(2\pi)^5} \frac{k_a k_b}{k_i} \left(|T_{dir}|^2 + |T_{exc}|^2 + |T_{dir} - T_{exc}|^2 \right) \quad (1)$$

where \vec{k}_i , \vec{k}_a , and \vec{k}_b are the wave vectors for the initial, scattered and ejected electrons, and T_{dir} is the direct scattering amplitude given by:

$$T_{dir} = \left\langle \chi_a^-(\vec{k}_a, \mathbf{r}_1) \chi_b^-(\vec{k}_b, \mathbf{r}_2) C_{scat-eject} | V - U_i | \phi_{DY}^{OA}(\mathbf{r}_2) \chi_i^+(\vec{k}_i, \mathbf{r}_1) \right\rangle \quad (2)$$

where r_1 and r_2 are the coordinates of the incident and the bound electrons, χ_i, χ_a , and χ_b are the distorted waves for the incident, scattered, and ejected electrons respectively, and $\phi_{DY}^{OA}(r_2)$ is the initial bound-state Dyson molecular orbital averaged over all orientations. The factor $C_{scat-eject}$ is the Coulomb interaction between the two final state electrons. We have found that using the exact interaction can over-estimate the strength of the interaction for atoms so we have used the Ward-Macek approximation [18] for atoms and the exact interaction for molecules. The initial state interaction potential between the incident electron and the neutral molecule is V , and U_i is a spherically symmetric approximation for V . This potential is called the distorting potential and it is used to calculate the initial-state distorted wave for the incident electron $\chi_i^+(\vec{k}_i, \mathbf{r}_1)$. For the exchange

amplitude T_{exc} , particles 1 and 2 are interchanged in equation (2). The Schrödinger equation for the incoming electron wave-function is given by:

$$(T + U_i - \frac{k_i^2}{2})\chi_i^+(\vec{k}_i, r) = 0 \quad (3)$$

where T is the kinetic energy operator and the '+' superscript on $\chi_i^+(\vec{k}_i, \mathbf{r})$ indicates outgoing wave boundary conditions. The initial state distorting potential contains three components $U_i = U_s + U_E + U_{CP}$, where U_s contains the nuclear contribution plus a spherically symmetric approximation for the interaction between the projectile electron and the target electrons which is obtained from the quantum mechanical charge density of the target. For H_2 , the charge density is $2|\phi_{DY}|^2$ (the 2 is for double occupancy and the original non-averaged Dyson orbital is used). For N_2 , the charge density is summed over the seven occupied orbitals. The nuclear contribution to U_s is the interaction between the projectile electron and the 2 nuclei averaged over all orientations which means that the net charge of the two nuclei is placed on a thin shell whose radius is the distance of the nuclei from the center-of-mass.

The potential U_E is the exchange potential of Furness-McCarthy [19] which approximates the effect of the continuum electron exchanging with the passive bound electrons in the molecule, and U_{CP} is the correlation-polarization potential of Perdew and Zunger [20] (see also Padial and Norcross [21]).

The final state distorted waves are obtained the same as the initial state except that the final state charge density for the ion is used to calculate U_s . The final state charge density is obtained the same as the initial state except that the occupancy number for the active electron is unity.

4. Results and Discussion

In a previous paper, we examined the TDCS for electron-impact ionization of the H_2 molecule in comparison to atomic He. We found that there was an overall good agreement between experiment and theory [9]. Both experiment and theory predict a much more complicated interference pattern, particularly in the binary peak region, than is given by the elementary Cohen-Fano interference factor, I^{CF} . We also found that the I-factor is more sensitive to projectile angular scans than to ejected-electron energy scans which indicate that for the present set of kinematics the diffraction of the projectile from two scattering centers is more important than the interference between electron waves emitted from two different centers.

In this work, we have extended our TDCS measurements to N_2 molecules for the same kinematics. Figure 2 compares H_2 and N_2 results for 250 eV incident electron impact, fast electron scattering angle of 15° and ejected electron of 50 eV. The top two panels of the figure contain the previously presented results for H_2 and He. On the left are the TDCS for both the atom and molecule normalized to unity at the TDCS peak and the right panel contains the I-factors. The experimental I-factor is the ratio of the H_2 and He (not shown) TDCS on the left, the red solid curve on the right is the ratio (I^{M3DW}) of the M3DW H_2 and He TDCS shown on the left and the blue dashed curve is I^{CF} (also sometimes called the Stia factor).

While the elementary Cohen-Fano factor exhibits qualitatively the shape of the I-factor, both the experimental and M3DW I-factors indicate a more complicated structure and the M3DW is in reasonable qualitative agreement with experiment. Results for N_2 and N are presented in the bottom 2 panels. The left panel presents theoretical and experimental results for N_2 as well as theoretical results

for atomic N. All the TDCS have been normalized to unity at the peak. The lower right panel contains the corresponding I-factors. Since there were no experimental TDCS available for atomic N, the experimental I-factors for N₂ represent the experimental N₂ TDCS divided by the theoretical atomic N TDCS. Again it is seen that the experimental and M3DW I-factors have a more complicated structure than I^{CF}. While the I-factors for H₂ and N₂ are similar, the I-factor for H₂ exhibits a double peak structure within the angular range of binary peak which is supported by the experimental data while N₂ has only a single peak in this angular range with a much smaller peak outside the angular range of the experiment. The M3DW I-factor for N₂ is in very good agreement with experiment for all the measured points.

The logic behind the I-factor is that dividing the molecular TDCS by the atomic TDCS removes all atomic effects in the TDCS and leaves two-center (molecular) effects. The Cohen-Fano I^{CF} factor shows what this interference is predicted to look like for photon impact where the interference would result from the target electron being ejected from two centers. The fact that the experiment and theory qualitatively have the shape of the Cohen-Fano factor indicates that this type of interference is present. The fact that the actual structure is much more complicated indicates that the other two possible modes of interference (i.e. incident electron being diffracted from two scattering centers and the scattered electron wave being emitted from two centers) is more important.

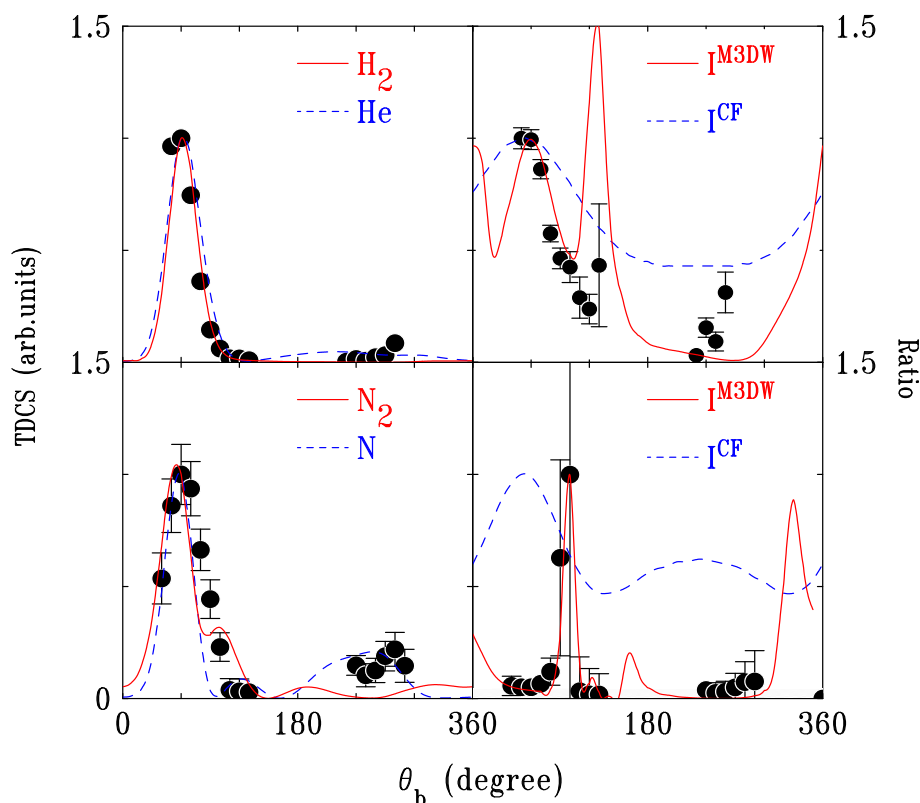


Figure 2. TDCS and Interference factors for 250 eV electron impact ionization of H₂ and N₂ as a function of the ejected electron angle θ_b . The fast final state electron scattering angle is 15° and the ejected electron energy is 50 eV. The top two panels of the figure contain results for H₂ and He (TDCS on left and TDCS ratios on right) and the bottom two panels contain results for N₂ and N. Solid circles-present data; solid red curve on the left - M3DW TDCS for H₂ and N₂; dashed blue curve on the left - M3DW TDCS for He and N; solid red curve on the right - I^{M3DW}; dashed blue curve on right I^{CF}. See text for more information.

In summary, we see that there is significant interference at the quantum level that it is not amenable to a simple classical interpretation for lower energy incident electrons. These results demonstrate for the first time that Young's type double slit interference effects are present for ionization of N_2 as well as H_2 . These are preliminary results. We are in the process of measuring more TDCS for N_2 at other projectile scattering angles and ejected electron energies and will present a more complete set of results in the near future.

Acknowledgments

This work was supported by the Scientific and Technological Research Council of Turkey (TUBITAK) through grant 109T738. The theoretical work of HC and DM was supported by US National Science Foundation under Grant. No. PHY-1068237.

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