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Observation of two-center interference effects for electron impact ionization of N_2

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Abstract

In 1966, Cohen and Fano (1966 *Phys. Rev.* **150** 30) suggested that one should be able to observe the equivalent of Young's double slit interference if the double slits were replaced by a diatomic molecule. This suggestion inspired many experimental and theoretical studies searching for double slit interference effects both for photon and particle ionization of diatomic molecules. These effects turned out to be so small for particle ionization that this work proceeded slowly and evidence for interference effects were only found by looking at cross section ratios. Most of the early particle work concentrated on double differential cross sections for heavy particle scattering and the first evidence for two-center interference for electron-impact triple differential cross section (TDCS) did not appear until 2006 for ionization of H_2. Subsequent work has now firmly established that two-center interference effects can be seen in the TDCS for electron-impact ionization of H_2. However, in spite of several experimental and theoretical studies, similar effects have not been found for electron-impact ionization of N_2. Here we report the first evidence for two-center interference for electron-impact ionization of N_2.

Keywords: electron impact ionization, two-center interference, N_2

(Some figures may appear in colour only in the online journal)

Introduction

The concept of wave–particle duality is considered a milestone in the development of quantum mechanics. The observation of interference fringes from coherent light passing through two closely spaced slits became the basis for the modern wave theory of light. These early studies for photons helped establish the foundations of interference phenomena as a fundamental signature for quantum ideas and subsequent interference experiments for particle impact were carried out using several particles including electrons, neutrons and heavy species such as bare carbon ions and Kr ions [1–3].

The idea of interference in collisions of diatomic molecules with photons was first discussed by Cohen and Fano [4] in 1966. Based upon the wave–particle duality, one would expect that effects similar to those seen for photons should also be seen for particle impact. Most of the early particle work concentrated on double differential cross sections and the first experimental evidence for double-slit interference effects in single ionization of molecules by ion impact was presented by Stolterfoht *et al* [3] in 2001. In 2002, Stia *et al* [5, 6] suggested that Cohen–Fano interference effects should also be expected for electron impact ionization of H_2. The first evidence for electron–H_2 interference was reported by Milne-Brownlie *et al* [7] in 2007 by looking at the relative sizes of the binary and recoil peaks in the coplanar triple differential cross section (TDCS) for electron-impact ionization of H_2. This observation was subsequently confirmed for different kinematics by Casagrande *et al* [8].

In the Cohen–Fano model, the incident projectile is a photon. Consequently, the only two-center interference physics contained in the model for the ejected electron is emission from

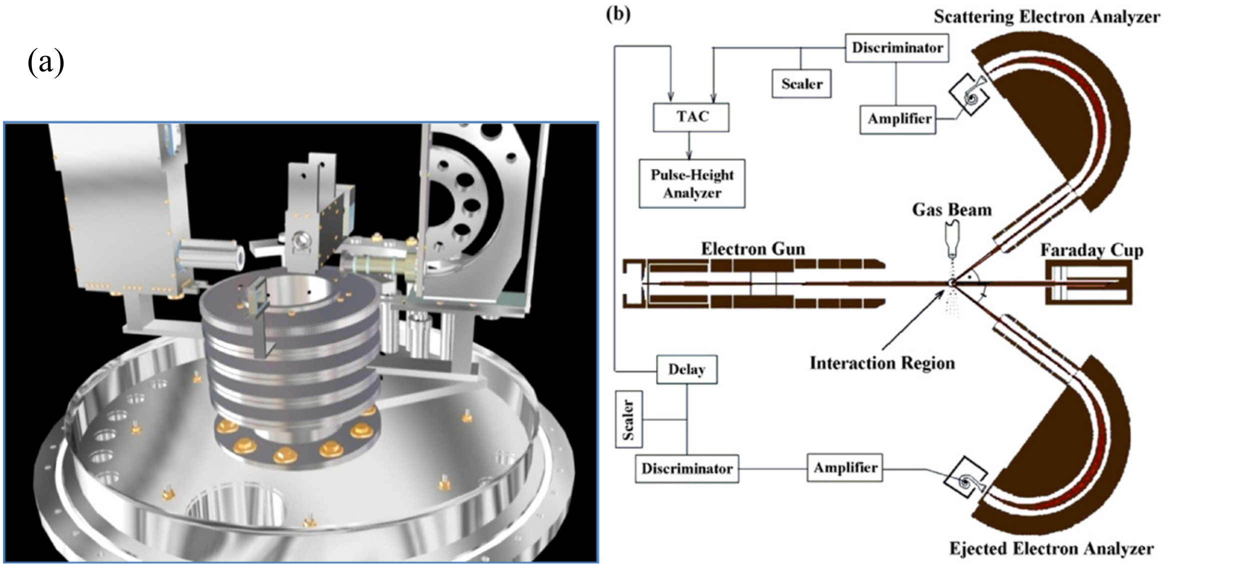

Figure 1. (a) Sketch of electron spectrometer. The main components are: electron gun, two electron analyzers and a Faraday cup and (b) coincidence electronics used to accumulate the coincidence timing spectrum at each set of kinematics.

two different scattering centers (i.e. the two slits in the Young's experiment). However, for incident electrons, there are (at least) three different possible two-center interference effects: (i) incident electron being diffracted by two scattering centers; (ii) scattered electron being emitted from two centers; and (iii) ejected electron wave being emitted from two centers. Madison and coworkers [9, 10] examined the three different types of possible two-center interference effects for electron-impact ionization of H_2 using the molecular three-body distorted wave (M3DW) approximation. Since the model of Stia *et al* [5] is based upon the Cohen–Fano approach, the only two-center interference effects in this model is also the ejected electron being emitted from two nuclei. The M3DW calculations for H_2 [9, 10], on the other hand, contain all three possible two-center interference effects and model calculations indicated that the most important contribution to two-center interference is coming from the diffraction of the incident projectile from two scattering centers.

As mentioned above, looking at the ratio of binary to recoil peaks provides indirect evidence for two-center interference effects at the molecular level. A different approach for finding Young's two-center interference effects for H_2 was recently reported by our group. Cohen–Fano noted that the best way to look for double-slit interference effects was to look at the ratio of the molecular H_2 cross section to the atomic H cross section. The logic is that this ratio, called the interference factor (*I*-factor), should contain only the two-center effects since the single center effects should cancel. Due to the difficulty of measuring atomic H cross sections, we looked at the ratio of molecular H_2 to He cross sections both experimentally and theoretically and we found a rich structure in both the experiment and theory. This structure was interpreted as a direct observation of two-center interference effects and very nice agreement between experiment and theory was found [9, 10]. The Cohen–Fano *I*-factor [4] $\left(I^{\mathrm{CF}}\right)$ (same as the Stia *et al* [5] *I*-factor) was only in very rough qualitative agreement with experiment and I^{CF} did not predict any of the detailed structure which indicates that the full two-center interference effects are much more complicated that just the double-slit component.

If two-center interference effects are present for H_2, then one would expect that they should also be seen for N_2. However, in spite of several searches, no conclusive evidence for two-center interference effects have been found for N_2 (11–16). In a theoretical study of low incident-energy electron-impact ionization of the N_2 ($3\sigma_g$) molecular state, Gao *et al* [11] predicted strong Young's double slit type interference effects for highly asymmetric scattering for coplanar 180° (back scattering). Murray *et al* [12, 13] performed experiments on the $3\sigma_g$ and $3\sigma_u^*$ states of N_2 in a coplanar asymmetric geometry and the predicted interference peak was outside the experimentally accessible angular range. Several other studies of TDCS for electron-impact ionization of N_2 have been performed [14–16] and none of them found any evidence for two-center interference for N_2.

In this paper, we report a study looking for evidence of two-center interference in (e, 2e) ionization of N_2 by looking at the *I*-factor which, in this case, is the ratio of the molecular N_2 cross sections divided by the atomic nitrogen N cross sections. Similar to the H_2 study, we do not have experimental data for ionization of atomic N. In this case, we use theoretical N cross sections calculated in the M3DW as the denominator for both experiment and theory (as has been routinely done for heavy particle scattering [2, 3]). In this paper, TDCS measurements and ratios are presented for ionization of the $3\sigma_g$ valance molecular orbital of N_2 in the intermediate-energy range and very strong interference effects are found. A preliminary report of this work was recently published in a conference series [17].

Experiment

This study has been conducted in a conventional (e, 2e) spectrometer (see in figure 1(a)) which has been well documented in previous works [18, 19] and so will only be briefly described here. The spectrometers in electron collision laboratory (e-COL) have been used to measure TDCSs for electron impact ionization of He, Ar and H_2 [9, 10, 20–23]. A vacuum pressure of $\approx 8.10^{-8}$ mbar is achieved. The magnetic field in the collision region is reduced to about 3 mG by using μ-metal shielding as well as the Helmholtz coils that eliminate the Earth's magnetic field. The electron gun consists of a tungsten filament, and 7 element electrostatic lenses including electrostatic deflectors allowing the beam to be focused onto the 2 mm diameter interaction region. The incident electron beam energy can be varied from 40 to 350 eV. Typical electron currents are around 1–3 μA and the electron current remains stable over a long time. It is essential for the (e, 2e) technique to obtain accurate knowledge of the energies of the incident, scattered and ejected electrons. Scattered and ejected electrons are determined by two hemispherical electrostatic analyzers. Each analyzer consists of a five element input electrostatic lens system and a Channeltron (CEM). The (e, 2e) technique has an advantage for identifying single ionization events for which the outgoing electrons are originated from the same ionization event. Using standard coincidence timing techniques, the arrival times of the electrons detected in each analyzer were used to determine if the electrons originated from the same ionization event. Coincidence electronics are shown in figure 1(b).

The results for ionization from $3\sigma_g$ orbital of N_2 presented in this paper were collected in a coplanar asymmetric geometry, where the scattered and ejected electrons are detected on the same plane.

The incident electron current was around 3 μA. In this study the obtained binding energy resolution was $\approx$1.4 eV (FWHM) for an incident electron energy $E_0 = 250$ eV, with the scattered electron being detected in coincidence with an ejected electron with $E_b = 50$ eV.

Theory

We have used the M3DW approximation to calculate the TDCS for N_2 and the atomic 3-body distorted wave (3DW) approximation to calculate the TDCS for N. The theory for these calculations has been presented elsewhere [11, 25, 26] so we will not repeat the equations here. However, we should note that we have used the orientation-averaged molecular orbital approximation [24] which was shown to give very good agreement with experimental TDCS data for H_2. For the N calculation, we have used Hartree–Fock bound state wavefunctions and for N_2, we have used wavefunctions calculated using density functional theory. Finally, we have used the exact final state electron–electron interaction (normally called post-collision-interaction or PCI).

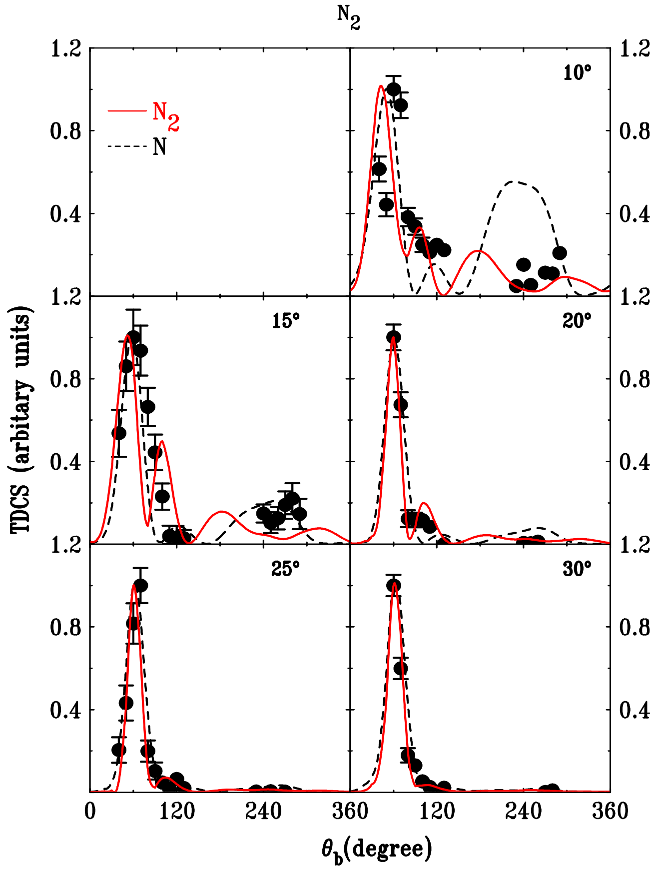

Figure 2. TDCS for 250 eV electron impact ionization of the $3\sigma_g$ valance state of N_2 and atomic N as a function of the ejected electron angle θ_b. The projectile scattering angle is noted in each sub-figure from 10° to 30° in steps of 5°. Solid circles—N_2 experimental data, solid (red) curve—M3DW calculations for the N_2 molecule and dashed (black) curve—3DW calculations for the nitrogen atom.

Results and discussion

In previous papers, we compared experimental and theoretical interference factors (*I*-factors) for electron-impact ionization of the H_2 molecule [9, 10]. We found that the *I*-factor exhibited a very complicated structure and, overall, there was very good agreement between theoretical cross sections and experimental data. The observed theoretical and experimental two-center interference factor exhibited significantly more structured than the double-slit Cohen–Fano interference factor (I^{CF}). We found that interference is more sensitive to the projectile scattering angle than the ejected electron energy and we found that projectile diffraction from two scattering centres is more important than the ejected electron being emitted from the two different centers [9].

Figure 2 compares the experimental and theoretical triple differential cross sections for 250 eV electron impact ionization of N_2 and N for fixed projectile scattering angles ranging from 10° to 30° by steps of 5° and a fixed ejected electron energy of 50 eV. For the smaller projectile scattering angles, theoretical binary peaks are slightly shifted to the lower ejected angles compared with the experimental data. However, the agreement with experiment improves for increasing

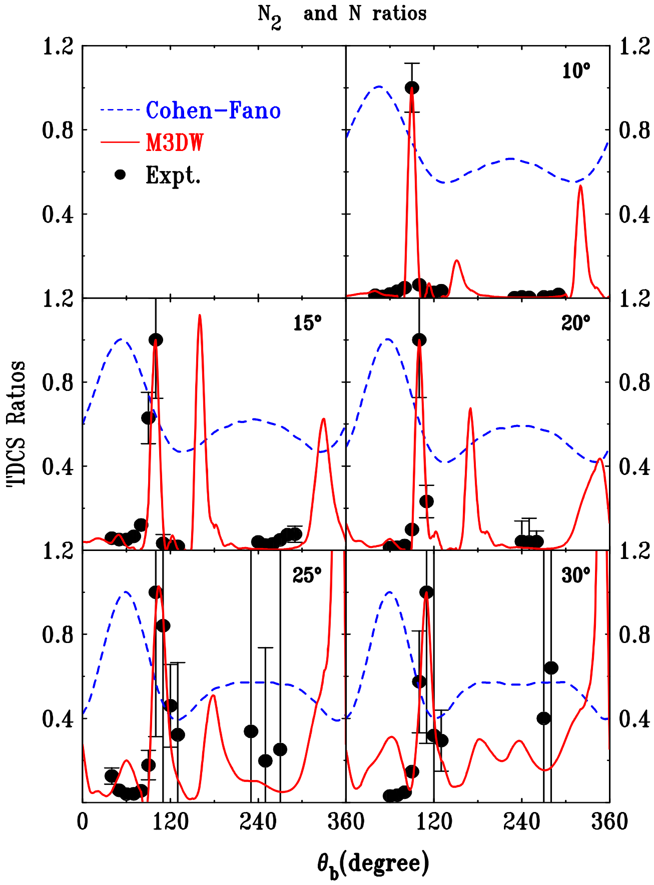

Figure 3. Interference factor for 250 eV electron impact ionization of the N_2 molecule as a function of the ejected electron angle θ_b. The projectile scattering angle is noted in each sub-figure from 10° to 30° in steps of 5°. Solid circles—experimental data, solid (red) curve—M3DW, and dashed (blue) cuve—I^{CF}.

projectile scattering angle and quite good agreement is found for the largest angles. The recoil peak intensity decreases with increasing projectile scattering angle, which is seen in both the theoretical and experimental data.

Theoretical calculations for N_2 consistently predict a shoulder in the binary region around 100°, with the intensity of the shoulder decreasing with increasing projectile scattering angle from 15°. Although this shoulder is not seen in the experimental data, there is a small suggestion for the possibility of a shoulder at 10° and also perhaps at 25°. Overall, the theoretical calculations for N_2 are in reasonably good agreement with the experimental data.

Figure 3 compares the experimental and theoretical *I*-factors (ratios of molecular to atomic cross sections). Since experimental atomic N cross sections are not available, we used theoretical N cross sections instead which is the norm for heavy particle scattering. We normalized both the experimental and theoretical interference factors to unity at the low angle maximum. Also included in figure 3 are the Cohen–Fano *I*-factors (which are the same as the Stia *et al* *I*-factors).

For the low-angle peak, both the M3DW results and experiment predict the peak at the same angle and they are in an excellent agreement. Theory predicts two more peaks—one around the 150° and one in the 330°–360° range, both of which are inaccessible to experiment although there is a suggestion for the high-angle peak at large projectile scattering angles.

The Cohen–Fano *I*-factor I^{CF} predicts a broad peak for small ejected-electron angles with the maximum occurring at significantly smaller angles than was found in either the present experiment or theory. For the larger ejection angles, I^{CF} predicts a very broad small peak which is also not found in either the experiment or theory. For the H_2 molecule, we found a qualitative agreement between our results and I^{CF}. However, for the N_2 molecule, there is little similarity between I^{CF} and the present results which indicates that the three different possible two-center effects yield a much more complicated interference pattern that the single double-slit possibility.

Gao *et al* [11], found evidence for a strong interference effect for N_2 in the scattering plane for electron emission at 180° Andrew Murray's group looked for interference effects for N_2 [13] and they did not find any. In this work, there is also a theoretical suggestion for interference effects near 180° (which is not experimentally accessible). On the other hand, the excellent agreement between experiment and theory found for the main peak around 100° represents the first direct evidence for two-center interference effects in electron-impact ionization of N_2. One might think that the theoretical peak results from the (probably) unphysical shoulder on the binary peak. However, we checked and this is not the case. There is no obvious shoulder in the experimental data and it has a peak at exactly the same angle as the theory. This peak is more strongly influenced by the shape of the atomic N cross sections than the molecular N_2 cross sections. The weakness of this approach is that the same atomic cross sections are used for both experiment and theory and it would be much better to have experimental cross sections as we did for H_2. Nevertheless, we think that the good agreement between experiment and theory is significant and represents the first evidence for interference effects for N_2.

Conclusions

We compared experimental and theoretical (e, 2e) cross sections and *I*-factors for 250 eV electron-impact ionization of the N_2 molecule in the scattering plane. We found reasonably good agreement between the theoretical M3DW TDCS results and experiment. However comparing experiment and theory for the TDCS does not provide a very good method for identifying two-center interference effects since it is not clear how these effects are manifested in the cross sections. In 1966, Cohen–Fano [4] noted that a better test is to take ratios of the TDCS for the molecule divided by the TDCS for the corresponding atom (the *I*-factor). The logic was that dividing by the atomic cross sections would remove single center effects and leave only two-center effects.

Evidence for two-center interference effects have now been demonstrated for electron–H_2 scattering [7–10]. Although there were several experimental attempts to find two-center interference effects for N_2, no experimental evidence has been found in prior work. In this work, we compared the theoretical and experimental *I*-factors for N_2 and found a strong peak within the angular range of the binary peak and the theoretical and experimental results were in excellent agreement with each other. This observation represents the first evidence for two-center interference effects to be seen for N_2. The *I*-factor represents a better test for interference than looking directly at the TDCS since it is not clear how interference effects will be manifested directly in the cross sections. Previous works for N_2 did not look at the *I*-factor and this is the reason they did not see any evidence for two-center interference.

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