Interference effects in electron emission spectra for 3 MeV/u H\(^+\) + O\(_2\) collisions

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Abstract. The present work extends the study of oscillatory structures associated with electron emission from diatomic molecular targets to 3 MeV/u H\(^+\) + O\(_2\). Electrons ejected from O\(_2\) were detected in the energy range 5 to 400 eV and for observation angles 30º, 60º, 90º and 150º. Experimental molecular O\(_2\) cross sections normalized to theoretical molecular O\(_2\) cross sections reveal oscillatory structures suggestive of secondary interferences as evidenced by their independence on the observation angle. Contrary to results for 1, 3 and 5 MeV/u H\(^+\) + H\(_2\), no obvious evidence for primary Young-type interferences was seen, a result similar to that found for H\(^+\) + N\(_2\) collisions.

1. Introduction
Quantum mechanical interference effects are a well-known consequence of the wavelike behavior of particles, and have generated considerable interest among experimentalists and theoreticians for decades. Early predictions and observations of two-center interference effects in the photoionization of N\(_2\) and O\(_2\) [1] and of H\(_2\) [2] were followed about thirty years later by detailed theoretical investigations of similar phenomena for photon collisions with H\(_2^+\) [3]. In the latter work the molecular orbitals are represented by a combination of two atomic orbitals, the continuum electron is described by a plane wave, and the nuclei do not move (fixed-in-space molecules). Moreover, experiments involving collisions of ions and electrons with H\(_2\) [4,5,6,7,8,9,10,11] uncovered structures analogous to the interferences observed in Young’s famous two-slit experiment. Corresponding theoretical calculations H\(_2\) [12,13,14,15] were generally in good agreement with the experimental measurements. In addition to Young-type interferences, more detailed analysis of the measured electron spectra in [6,9] revealed second-order oscillations superimposed on the primary interference structures with about 2-3 times higher frequency.

Recently, the work of Hossain et al. [9] for 1-5 MeV/u H\(^+\) + H\(_2\) was extended to proton collisions with more complex diatomic molecular targets. In contrast to the Young-type and secondary oscillatory structures identified in the ejected electron spectra of H\(_2\), results for 1-5 MeV/u H\(^+\) + N\(_2\)
collisions revealed apparently only secondary oscillations [16]. Similar findings were recently reported for 1.9 MeV/u O$^{3+}$ + O$_2$ [17].

In order to gain new insight into the interference patterns reported for the above mentioned collisions, experimental investigations have been performed for 3 MeV/u H$^+$ + O$_2$. As a result of the larger internuclear separation of 2.28 a.u. for O$_2$, compared with 2.1 a.u. for N$_2$ and 1.4 a.u. for H$_2$, the primary Young-type interference structures would be expected to have higher frequencies. Additionally, due to the similar projectile energies but different molecular targets, the results for H$^+$ + O$_2$ collisions allow quantitative comparison with previous results for H$^+$ + N$_2$ [16].

2. Theory

Theoretical cross section calculations using different models [12,13,14,15,18] follow quite closely the general behavior of the experimental findings, and contribute to the interpretation of interferences resulting from coherent electron emission from the identical molecular centers of H$_2$. A description of the observed oscillatory structures is offered in the work of Ref. [4] by the relation:

$$ (\sigma_{H_2})_{norm} = \frac{d^2\sigma_{H_2}}{d\Omega d\varepsilon} / \frac{d^2\sigma_{2H}}{d\Omega d\varepsilon} = \int \frac{d^3\sigma_{2H}}{dqd\Omega d\varepsilon} \left[ 1 + \frac{\sin|k - q|d}{|k - q|d} \right] \times dq / \int \frac{d^3\sigma_{2H}}{dqd\Omega d\varepsilon} dq $$

where $\sigma_{H_2}$ is the cross section for two-center emission, $\sigma_{2H}$ the cross section for emission from H, $\Omega$ is the solid angle, and $\varepsilon$ the energy of the outgoing electron. The sinusoidal term, where $|k - q|$ is the difference between the outgoing electron momentum $k$ and the momentum transfer $q$, and $d$ is the internuclear distance, describes the interference caused by coherent emission from the two nuclei.

For more complex systems, despite the difficulties involved in providing accurate theoretical calculations, efforts proved to be successful in identifying interference effects for 1, 3 and 5 MeV/u H$^+$ + N$_2$, (see Ref.[16]). In this latter work, the theoretical atomic N cross sections were calculated using a first-order impact parameter method. Hartree-Fock wavefunctions were used to describe the initial state of the N atom, while that corresponding to the ejected electron has been calculated in the field of the residual ion. In the present work, motivated by the results for H$^+$ + N$_2$ [16], theoretical calculations for molecular O$_2$ cross sections were performed using one-center wavefunctions, following the methods described in Ref. [19].

3. Experimental procedure

The measurements were performed at Western Michigan University using the 6-MV tandem Van de Graaff accelerator. A collimated beam of 3 MeV/u H$^+$ was directed into the scattering chamber onto an O$_2$ gas target supplied by a gas nozzle with the flow rate set to maintain a pressure of ~ 4 x 10$^{-5}$ Torr inside the chamber, for which single collision conditions were found to prevail. The incident beam intensity was measured downstream in a Faraday cup. Electrons emitted from the target were energy analyzed using an electrostatic parallel-plate spectrometer and counted with a channel-electron multiplier for ejection energies 5-400 eV and observation angles of 30°, 60°, 90° and 150° with respect to the incident beam direction. Electron spectra were measured without the target gas at a background pressure of ~ 2 x 10$^{-6}$ Torr to correct for residual gas events. Shielding with a μ-metal liner was used to minimize stray magnetic fields inside the scattering chamber.

4. Results and discussion

Following corrections for contributions from the background gas the ejected electron yields, which are proportional to the cross sections, were normalized to the calculated theoretical cross sections for 3 MeV/u H$^+$ + O$_2$ by setting the experimental value equal to the theoretical cross section near the middle of the spectrum (on a logarithmic scale) as shown in Figures 1a, b. As the theoretical cross sections decrease faster than the experimental yields for all electron observation angles, the
Experimental-to-theoretical O$_2$ cross section ratios exhibit rising trends as shown in Figures 1c,d, where the ratios are plotted as a function of the ejected electron velocity $k$ (results are shown only for two of the four measured angles). A similar behavior was observed in the work of Refs. [4,16].

The overall increase of the above mentioned ratios was removed by fitting a straight line to the ratios, clearly revealing the oscillatory structures as shown in Figure 2. The modified ratios have statistical fluctuations associated with experimental uncertainties that tend to become larger, at backward angles and higher electron velocities. The frequencies of these structures show no obvious dependence on the observation angle contrary to the behaviour expected for Young-type interferences [4,5,9]. To quantify the oscillations, a phase shifted sinusoidal function was used to fit the ratios:

$$f(k) = A\sin(ckd - w) + B;$$  \hspace{1cm} (2)

a method that was first used in Ref. [6]. In Equation 2 $d$ is the internuclear distance (2.28 a.u.), $c$ is an adjustable frequency parameter, $w$ is an adjustable phase shift, and $A$ and $B$ are constants. The resulting frequency $c$ and phase shift $w$ parameters are shown in Figure 3. The values obtained for 1.9 MeV/u O$^{5,8^+}$ + O$_2$ [17] are added for comparison. The frequency parameter $c$ remains nearly constant for all electron observation angles, a characteristic that is suggestive of secondary interferences [6,9], and was also seen for 1-5 MeV/u H$^+$ + N$_2$ [16] and 1.9 MeV/u O$^{5,8^+}$ + O$_2$ collisions [17].

With an apparent minimum at 90º, the phase parameter $w$ displays a similar pattern to that reported for H$^+$ + N$_2$ [16] and O$^{5,8^+}$ + O$_2$ [17]. Notably, an oscillation interval of $\Delta k \sim 4$ a.u. is found as in Ref. [17], a value two times larger than for 3 MeV/u H$^+$ + N$_2$ ($\Delta k \sim 2$ a.u.). The reason for this difference is presently not known but may be due to the differences in the N$_2$ and O$_2$ wavefunctions.
5. Conclusions
Evidence for interference effects in the electron emission spectra of O$_2$ by 3 MeV/u H$^+$ impact is revealed. In qualitative agreement with the results obtained for H$^+$ + N$_2$ [16], the observed oscillatory structures are independent of the observation angle, and consequently are identified as second-order interferences from intramolecular scattering. In contrast with the oscillatory structures reported for H$^+$ + H$_2$ [9], no primary Young-type interferences are seen suggesting that interference effect for heavy targets differ fundamentally from those for H$_2$. To better understand these differences, theoretical calculations are needed to provide a quantitative understanding of the frequency and magnitude of the secondary oscillations.

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