Nuclear Instruments and Methods in Physics Research B 267 (2009) 201–205

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Interferences in coherent electron emission from diatomic molecules

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ARTICLE INFO

Available online 17 October 2008

PACS:
34.50.Fa
32.80.Fb

ABSTRACT

Recent studies of electron emission from molecular hydrogen by the impact of fast ions have shown the existence of interference effects. The interferences are manifested as oscillations in the velocity (or energy) distributions of the ejected electrons, and are analogous to the interference of light in Young's two-slit experiment. The frequencies of the oscillatory structures depend strongly on the electron observation angle and to a lesser extent on the collision velocity. Additionally, secondary oscillations with ~2–3 times higher frequencies attributed to scattering of the primary electron "wave" at the other atomic center are found to be superimposed on the primary oscillations. More recently, electron interference studies have focused on diatomic molecules more complex than H_2, including N_2 and O_2, for which only structures due to secondary interferences are apparently observed. Here, these various results are reviewed, outstanding questions identified, and future directions indicated.

1. Introduction

The observation [1] of oscillatory structures in the ejected electron energy spectra of H_2 impacted by fast ions confirmed the long-standing prediction [2] of Young-type interferences resulting from the coherent emission of electrons from identical atomic centers. Representing a fundamental aspect of quantum mechanics [3], the interference structures demonstrated that coherent emission from atomic centers, corresponding to the "slits" in Young's experiment for the interference of light, can lead to outcomes significantly different from those observed for atoms. The initial observation carried out for 60 MeV/u $Kr^{34+} + H_2$ collisions [1] prompted substantial interest and led to numerous follow-up studies by several investigators [4–11]. Accompanying the experimental investigations, several theoretical formulations utilizing different approaches [12–15] have been developed to describe the features of the observed Young-type interference structures.

For fast ion collisions with H_2 [4–6,9–11] the relatively small oscillatory interference structures, which are superimposed on an exponentially decreasing "background" of continuum electron emission, exhibit a strong dependence on the ejected electron angle [4,5] and a lesser dependence on the collision velocity [5]. These dependences are in general agreement with the theoretical predictions [12–15] for forward electron ejection angles but significant discrepancies exist for backward angles. In addition to the primary Young-type interferences, secondary oscillations superimposed on the main interference structures have been observed [5,16]. These secondary oscillations, with ~2–3 times higher frequency, have been attributed to scattering of the primary electron "wave" at the second atomic center resulting in interference with the original wave [16]. Moreover, for $H^+ + H_2$ collisions some evidence for still higher frequency oscillations superimposed on the primary and secondary interference structures was observed [5]. High-frequency oscillations have also been seen in low velocity *atomic* collisions of 20 keV He^+ and 40 keV He^{2+} with atomic He [17], leading to speculation that these oscillations are due to coherent emission from the transient molecule formed during the collision of homonuclear partners [5], a hypothesis that still needs verification. These various interference characteristics associated with electron emission from H_2 have been reviewed by Tanis and Hossain [18].

More recently, electron interference studies have focused on diatomic molecules other than H_2, including 1–5 MeV $H^+ + N_2$ [19] and 30 MeV $O^{5,8+} + O_2$ [20]. In contrast to results for H_2, oscillatory structures in the ejected electron spectra that do not depend on either the electron ejection angle or the collision velocity are observed. This behavior, characteristic of secondary interferences due to intramolecular scattering, consequently suggests that primary Young-type interferences are strongly suppressed or nonexistent. In fact, theoretical calculations predict suppression of the primary interferences but not to the extent observed in the measurements [19].

While [18] is concerned with the behavior and characteristics of interferences in electron emission from H_2, the present manuscript contrasts the results for H_2 with those observed for more complex molecules having several orbitals. Here, results involving oscillatory structures in the ejected electron spectra of diatomic molecules will be reviewed, outstanding problems identified, and

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doi:10.1016/j.nimb.2008.10.011

future directions indicated. Due to space limitations, only studies in which the present author was involved will be presented and discussed.

2. Theory

The cross section for ionization of H_2 including coherent emission from the two centers leading to interferences in the ejected electron velocity distribution is described by the relation [1,3]:

$$\left(\frac{d^2\sigma_{H_2}}{d\Omega\, d\varepsilon}\right) = \int \frac{d^3\sigma_{2H}}{d\mathbf{q}\, d\Omega\, d\varepsilon}\left[1+\frac{\sin|\mathbf{k}-\mathbf{q}|d}{|\mathbf{k}-\mathbf{q}|d}\right] d\mathbf{q}, \quad (1)$$

where $d\Omega$ and $d\varepsilon$ are the solid angle and energy of the outgoing electron, respectively. The cross section σ_{H_2} denotes molecular two-center emission while σ_{2H} is the cross section for incoherent emission from the two H atoms. This latter cross section can be calculated from purely atomic cross sections or from a one-center molecular treatment. The sinusoidal term represents the interference caused by coherent emission from the two centers, where $|\mathbf{k}-\mathbf{q}|$ is the difference between the outgoing electron momentum $\mathbf{k}$ and the momentum transfer $\mathbf{q}$, and d is the molecular internuclear distance. This cross section is obtained after averaging over the random orientation of the internuclear H_2 axis (see [4]). To compare with experiment, Eq. (1) must be integrated over the momentum transfer $\mathbf{q}$.

To better reveal the relatively small oscillations expected for the interference structures superimposed on "backgrounds" of exponentially decreasing cross sections, the measured molecular H_2 cross sections are normalized to corresponding theoretical atomic H cross sections $d^3\sigma_{2H}/d\boldsymbol{q}\, d\Omega\, d\varepsilon$ for independent H atoms integrated over $\mathbf{q}$, i.e.,

$$\left(\frac{d^2\sigma_{H_2}}{d\Omega\, d\varepsilon}\right)_{norm} = \left(\frac{d^2\sigma_{H_2}}{d\Omega\, d\varepsilon}\right) \Big/ \left(\frac{d^2\sigma_{2H}}{d\Omega\, d\varepsilon}\right)_{theo}. \quad (2)$$

It is noted that in other formulations to reveal the oscillatory structure, the measured molecular cross sections have been normalized to measured atomic H cross sections [6], or ratios of the measured cross sections for complementary forward-backward angles have been examined for asymmetric effects [11].

In the theoretical work of Nagy et al. [12], an extended treatment of the interference term in Eq. (1) was formulated. In this model the interference structure is governed by the electron momentum component parallel to the beam direction, i.e., $k_\parallel = k\cos\theta$, with the result that the frequency of the oscillation decreases as the electron emission angle increases to 90°. According to this model, the cross section ratio given by Eq. (2) is approximated by

$$\left(\frac{d^2\sigma_{H_2}}{d\Omega\, d\varepsilon}\right)_{norm} = F\left[1+\frac{\sin\left[(k_\parallel - q_{min})d\right]}{(k_\parallel - q_{min})d}\right] + G, \quad (3)$$

where F and G ($F+G=1$) are the interfering and non-interfering cross section fractions, respectively, and the minimum momentum transfer $q_{min} = \Delta E/v$, with ΔE being the energy transferred and v the velocity of the projectile. Also, numerical integration of Eq. (1) over $\mathbf{q}$ using analytic Born approximation cross sections obtained from hydrogenic wave functions [21,22] leads to a varying oscillation frequency with observation angle. The Born calculations give results essentially identical with the corresponding ratios determined from CDW–EIS [13,15,23] calculations.

If q_{min} is small, Eq. (3) indicates that the experimental cross section ratios can be represented by the function [4]

$$f(k) = A\left[1+\frac{\sin(kcd)}{kcd}\right] + B, \quad (4)$$

where c ($\approx\cos\theta$) is an adjustable frequency parameter and A and B are constants ($A+B\approx 1$) representing the interfering and non-interfering contributions to the normalized cross section, respectively. In the case of second order oscillations, the argument of the sine function has been modified to $\sin(kcd - w)$ to allow for the possibility of a phase shift [16]. It is expected that formulations similar to Eqs. (1)–(3) also hold for more complex diatomic molecules such as N_2 and O_2, but to date this has not been verified.

3. Experimental method

For the results to be considered here, measurements for 1, 3, and 5 MeV H^+ were conducted at Western Michigan University using the tandem Van de Graff accelerator. The experimental setup has been described in [5]. Briefly, a well-collimated beam with intensity ~100–500 nA was directed into the scattering chamber onto an H_2 or N_2 target jet of diameter 2–4 mm obtained from a nozzle with the flow rate set to maintain a pressure of a $\sim 5\times10^{-5}$ Torr in the scattering chamber. Electrons emitted from the target were energy analyzed with a parallel-plate spectrometer and counted with a channel-electron multiplier for ejection energies ranging from about 2–300 eV and for angles in the range 30–150° with respect to the incident beam direction. The spectra were normalized to the incident beam intensity, and background spectra were recorded with no target gas to correct for residual gas events. Magnetic fields inside the scattering chamber were minimized by shielding with a μ-metal liner.

4. Results and discussion

4.1. Cross section ratios for H_2 and N_2

Here, we review recent electron interference results for H^+ ions colliding with H_2 and N_2. As noted in Section 2, to reveal the

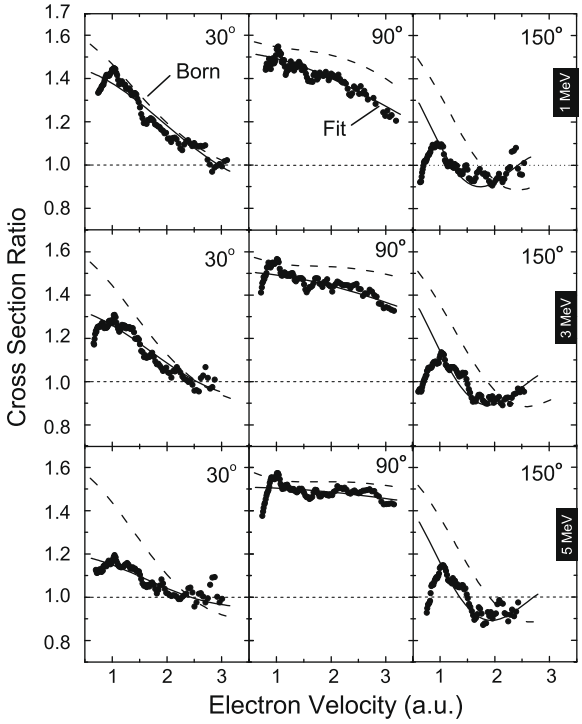

Fig. 1. Ratios of experimental molecular H_2 cross sections to theoretical atomic H cross sections, calculated in the CDW-EIS approximation [23], for electron emission by 1, 3, and 5 MeV H^+ impact on H_2 [5] for the electron observation angles indicated. The dashed curves are Born calculations from Eqs. (1) and (2), and the solid curves are fits to Eq. (4).