PHYSICAL REVIEW A **67**, 030702(R) (2003)

Interference effects in electron emission from H_2 by 68-MeV/u Kr^{33+} impact: Dependence on the emission angle

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(Received 27 August 2002; published 11 March 2003)

Angle- and energy-dependent cross sections for electron emission were measured for 68-MeV/u Kr^{33+} ions impacting on H_2. These results show, in accordance with our earlier observation, that interference effects are produced by the coherent emission of electrons from the two H atoms, in analogy with Young's two-slit experiment. Furthermore, the present results demonstrate that the observed oscillatory pattern varies with the electron observation angle, contrary to our earlier expectations but in agreement with recent theoretical predictions.

DOI: 10.1103/PhysRevA.67.030702 PACS number(s): 34.50.Fa, 32.80.Fb

Studies of particle-induced ionization have devoted particular attention to the molecular target H_2, which is the simplest molecule composed of two atoms. While the overall ionization is well understood [1–5], little is known about phenomena associated with the indistinguishability of the atomic H centers. In this case, the contributions to ionization from each center add coherently, and interference effects may be expected in the ionization spectra. Such electron emission from H_2 may be closely related to Young's two-slit experiment, which played an essential role in the early development of quantum mechanics.

In the past decades, effort has been devoted to reveal these interference effects in atomic collisions with H_2. Early studies of collisionally induced interferences from H_2 centered on the processes of electron capture [6] and photoionization [7]. These studies were followed by additional theoretical work (see Refs. [8,9] and references therein), whereas experimental work was limited [10]. Related experimental investigations with synchrotron radiation [11,12] focused on heavier molecules where one atomic center is photoionized (e.g., in an inner shell), followed by electron scattering at the other center. These scattering phenomena differ, however, from Young's experiment, where both slits simultaneously emit radial waves. The different cases are discussed by Messiah [13].

Recently, experimental evidence for interference effects has been found in H_2 electron emission spectra induced by very fast projectiles [14]. In that work, hereafter referred to as I, the analysis indicated that the use of a high projectile velocity is important because it enhances interference effects. The spectra obtained at forward angles (e.g., 30°) exhibited an oscillatory structure in good agreement with model calculations. Results obtained at backward angles (e.g., 150°) indicated a similar trend which, however, was not conclusive due to limited statistics. More recent data for 3- and 5-MeV $H^+ + H_2$ collisions also show evidence for interference effects in electron emission spectra [15].

The experimental work motivated various theoretical studies [16–19] that reveal detailed properties of the interference effects. It was recognized that dipole transitions and binary-encounter processes play different roles in the ionization process leading to interference [14,18]. In I, due to the high projectile velocity, model calculations focused on dipole transitions at forward angles, which seemed to indicate that the interference pattern is quite independent of the electron emission angle. However, at 90°, electron emission is strongly dominated by binary encounter collisions [20] and, consequently, the interference structures are expected to diminish. Moreover, a new theoretical work [17] shows explicitly that the frequency of the oscillation should decrease as the emission angle increases up to 90°.

In the present work, we provide decisive experimental evidence for interference effects in electron emission from H_2 resulting from 68 MeV/u Kr^{33+} ion impact. Specifically, measurements with improved statistics reveal pronounced oscillatory structures at forward and backward angles. Moreover, the measurements exhibit a previously unexpected dependence of the interference structures on the electron emission angle, indicating a varying oscillation frequency. This result is shown to be consistent with a new model prediction [17] and calculations based on the Born approximation.

In accordance with previous work [14,16,18], the cross section for electron emission from H_2 relevant for the present experiment is given by (atomic units are used throughout if not otherwise stated)

$$\frac{d\sigma_{H_2}}{d\Omega\, d\epsilon} = \int \frac{d\sigma_{2H}}{d\mathbf{q}\, d\Omega d\epsilon}\left[1 + \frac{\sin(pd)}{pd}\right] d^2 q_\perp , \tag{1}$$

where the solid angle $d\Omega$ and the energy $d\epsilon$ refer to the outgoing electron. The cross section $d\sigma_{2H}/d\mathbf{q}d\Omega d\epsilon$ describes incoherent electron emission from the two independent H atoms (denoted by the label 2H), but with mutually altered effective charges. The term in parentheses represents the interference caused by coherent emission from the two centers, where d is the internuclear distance of the H_2 molecule and $p = |\mathbf{k} - \mathbf{q}|$ is the difference between the electron momentum $\mathbf{k}$ and the momentum transfer $\mathbf{q}$.

Equation (1) is obtained after averaging over the random orientation of the internuclear H_2 axis. The presence of the interference term shows that the averaging procedure preserves the oscillatory features of the electron emission spectra. Moreover, Eq. (1) must be integrated over the momentum transfer vector $\mathbf{q}_\perp$ perpendicular to the beam direction. As discussed below, this integration was carried out using analytic cross sections $d\sigma_{2H}/d\mathbf{q}\, d\Omega\, d\epsilon$ for independent H atoms obtained from the Born approximation [5].

To investigate specific ionization mechanisms, in I the cross section was split into dipole and binary terms following the early work of Bethe [21]. The dipole term was derived using a peaking approximation, i.e., setting $q = 0$, whereas the binary encounter process contributes only a constant interference term. Thus, in I the integrated cross section was obtained as

$$\frac{d\sigma_{H_2}}{d\Omega\, d\epsilon} = \frac{d\sigma_{2H}^{dip}}{d\Omega\, d\epsilon}\left[1 + \frac{\sin(kd)}{kd}\right] + \frac{d\sigma_{2H}^{bin}}{d\Omega\, d\epsilon} s, \qquad (2)$$

where $d\sigma_{2H}^{dip}/d\Omega\, d\epsilon$ and $d\sigma_{2H}^{bin}/d\Omega\, d\epsilon$ are integrated cross sections referring to the dipole and binary parts, respectively, and $s \approx 1.6$ is a constant. Equation (2) suggests that the oscillatory structure in the cross section is produced solely by the dipole term. This term predicts a full sinusoidal oscillation when the product kd varies from 0 to 2π. With $d = 1.42$ a.u. for H_2, this oscillation is governed by the electron momentum k varying within the range of $0 - 4.3$ a.u. It is noted, however, that the dipole approach is limited primarily to forward and backward angles.

In an extended model, Nagy *et al.* [17] evaluated cross sections with an interference term that is governed by the momentum $p_\parallel = k_\parallel - q_\parallel$ parallel to the beam direction. This is plausible, since the $\mathbf{q}_\perp$ integration in Eq. (1) tends to diminish the perpendicular components due to averaging effects. Thus, it follows [17] that

$$\frac{d\sigma_{H_2}}{d\Omega d\epsilon} = \frac{d\sigma_{2H}^{int}}{d\Omega d\epsilon}\left[1 + \frac{\sin[(k_\parallel - q_{\min})d]}{(k_\parallel - q_{\min})d}\right] + \frac{d\sigma_{2H}^{non}}{d\Omega d\epsilon}, \qquad (3)$$

where $d\sigma_{2H}^{int}/d\Omega\, d\epsilon$ and $d\sigma_{2H}^{non}/d\Omega\, d\epsilon$ are, respectively, interfering and noninterfering parts of the cross section [17]. The component $k_\parallel = k \cos\theta$ is determined by the electron ejection angle θ, and $q_\parallel$ is equal to the minimum momentum transfer $q_{\min} = \Delta E/v_p$ obtained from the energy transfer ΔE and the projectile velocity v_p. For the high v_p used here, $q_{\min}$ is small. Thus, Eq. (3) predicts that the frequency of the oscillatory structure varies approximately with cos θ.

The experiments were performed at the Grand Accélérateur National d'Ions Lourds (GANIL), Caen, France. The scattering chamber and the electron spectrometer were the same as those used in I. A beam of 68 MeV/u Kr^{33+} ions with a current of $1-2$ μA was collimated to a size of about 2×2 mm^2 and directed onto an H_2 target of ~ 4 mm diameter obtained from a gas jet. Electrons emitted from the target were measured with a parallel-plate electron spectrometer for energies ranging from about 2 to 500 eV and for angles of 30°, 60°, 90°, and 150° with respect to the beam direction.

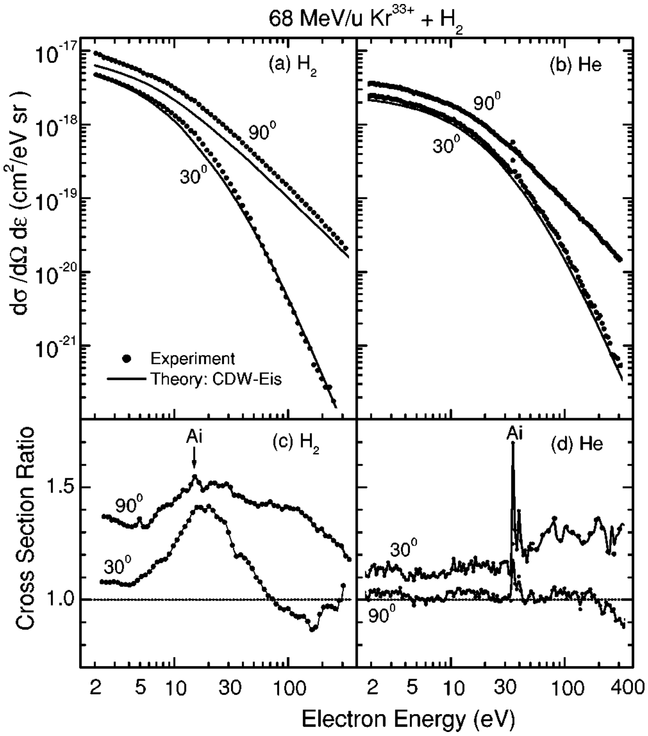

FIG. 1. Cross sections and ratios for electron emission by 68 MeV/u Kr^{33+} impacting on H_2 and He as a function of the ejected electron energy. In (a) and (b), experimental and theoretical cross sections are compared for H_2 and He, respectively, obtained at the observation angles 30° and 90°. In (c) and (d), cross-section ratios of experimental and theoretical results are given for H_2 and He, respectively. The peaks labeled "Ai" are attributed to autoionization.

Auxiliary measurements were performed with a He target as a benchmark for the H_2 results, and to put the cross sections for both targets on an absolute scale. This was done by normalizing the He cross section for 90° to theoretical results obtained by means of the continuum-distorted-wave eikonal-initial-state approximation (CDW-EIS) [22]. Earlier He ionization measurements with fast projectiles [23] have shown that the experimental data at 90° are well reproduced by these CDW-EIS calculations.

In Figs. 1(a) and 1(b), typical results for electron emission from H_2 and He, respectively, are shown for observation angles of 30° and 90°. Also plotted are theoretical CDW-EIS cross sections obtained for atomic targets. As in I, the He data were calculated using Hartree-Slater wave functions for the initial and final electron states [22]. The calculations for H were performed using an initial hydrogenic wave function with an effective target charge of $Z_T = 1.19$ in accordance with Ref. [18].

The cross sections are seen to vary strongly by several orders of magnitude with the electron energy [4,5]. Since the variation due to the interference term is less than a factor of 2, it is important to remove this strongly varying energy component. This is done by dividing the measured cross sections by the corresponding CDW-EIS cross sections. The results for 30° and 90° associated with H_2 and He are given in Figs. 1(c) and 1(d), respectively. The He data indicate a distinct peak labeled Ai near 33 eV produced by autoionization [23]. Apart from this peak structure, the He data show a smooth and nearly monotonic dependence on the ejected electron energy.

On the contrary, the 30° cross-section ratio for H_2 shows a non-monotonic behavior suggestive of an oscillatory structure. This structure is well outside the experimental uncertainties of the relative cross sections which are typically $\pm 10\%$. Larger uncertainties up to $\pm 30\%$ may exist for energies below about 5 eV, where the measured cross sections can be affected by spurious instrumental effects. A crucial point of the H_2 analysis is the possible contribution of autoionization electrons. In I, autoionization was estimated to contribute negligibly to the measured H_2 electron spectra. Nevertheless, we attribute the reproducible peak structure at about 13 eV to autoionization electrons (which, to our knowledge, have not been observed before in the electron emission from H_2). However, as outlined in I, the presence of the autoionization peak is not likely to alter the essential features of the interference pattern.

In Fig. 1(c), the H_2 cross-section ratios show an overall decrease with increasing energy. This decrease is contrary to the ratios obtained in I, which increased with energy. The difference is caused by the use of the effective charge $Z_T = 1.19$ for the initial state in the present work instead of $Z_T = 1.05$ in the earlier CDW-EIS reference calculations. We found that $Z_T = 1.19$ (due to a variational treatment [18]) better reproduces the nonoscillating behavior of the cross sections than does $Z_T = 1.05$ (which follows from the binding energy). Thus, the corrections to the cross-section ratios that were required in I are not needed here.

From Eq. (2), we recall that the interference term is governed by the momentum k (or velocity v) of the ejected electrons. Accordingly, we plot the cross-section ratios versus velocity v in Fig. 2, where results for different electron emission angles are given. Figures 2(a) and 2(d) show sinusoidal oscillations at forward and backward angles, respectively. The data for 30° are in good agreement with the previous measurements in I. From inspection of the results for the other angles, it appears that the frequency of the oscillatory structures varies with the electron emission angle.

To understand more quantitatively the interference structures, the experimental data are compared with theoretical results. Numerical calculations were performed using Eq. (1) in conjunction with analytical Born cross sections for atomic hydrogen [5]. These calculations were normalized to the corresponding integrated Born cross sections and are shown by the solid curves in Fig. 2. For all emission angles and velocities $v \gtrsim 1$, the Born results are found to be in excellent agreement with the calculations obtained from Eq. (3). Therefore, the results from Eq. (3) are not shown (keeping in mind that they are very close to the Born results).

To gain more information about the oscillation frequency, we used an analytical fit function based on the interference term that is common to Eqs. (2) and (3). This function has the form $A[1+\sin(kcd)/(kcd)]+B$, where A and B (with $A+B=1$) are interfering and noninterfering cross section fractions, respectively, and c is an adjustable frequency parameter. For simplicity, the fractions A and B were set equal to 0.5. (The Born calculations were based on similar fractions.) The actual values are of minor importance, since we are primarily interested in the oscillation frequency. The functions fitted to the experimental data are shown in Fig. 2, and the c values are given in Table I in comparison with $\cos\theta$. It is recalled from Eq. (3) that nearly equal values are expected for c and $\cos\theta$.

For the electron emission angles of 30° and 60°, there is excellent agreement between the experimental data and the normalized Born results as seen from Figs. 2(a) and 2(b).

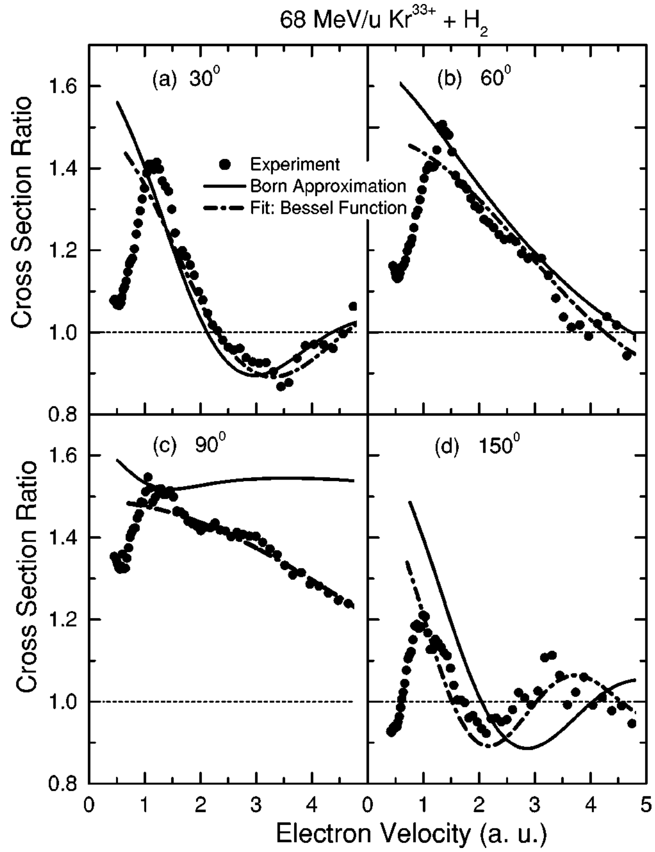

FIG. 2. Ratios of experimental to theoretical CDW-EIS cross sections plotted as a function of the ejected electron velocity for the electron observation angles indicated. The solid lines represent Born calculations from Eq. (1) and the dash dotted lines are obtained from fits to an analytic function (see text).

TABLE I. Fit values of the frequency parameter c as a function of the electron observation angle θ. The values of $\cos\theta$ are shown for comparison.

θ	$\cos\theta$	c
30°	0.87	0.96
60°	0.5	0.52
90°	0	0.29
150°	0.87	1.46

Accordingly, the experimental data are well reproduced by the analytic function with a frequency parameter c nearly equal to $\cos\theta$ (Table I). This finding supports the prediction of Eq. (3) that the interference term is governed by the momentum component $k_{\|}$ [17].

On the other hand, for the 90° ratio in Fig. 2(c), there is a significant deviation between the experimental results and the Born calculations. Consequently, the c value differs from $\cos\theta$ (Table I). For 90°, the models predict the cross-section ratio to be essentially constant. This constancy may be understood from the fact that electron emission at 90° is strongly dominated by binary-encounter processes [20]. Then, Eq. (2) predicts the normalized cross sections to be equal to $s\approx 1.6$, in agreement with the Born results. The experimental data decrease with velocity, however. Nevertheless, the experimental 90° data show the weakest dependence on the electron velocity in qualitative agreement with theory.

The experimental data for 150°, acquired with improved statistics compared to I, show an extended sinusoidal oscillation giving rise to a distinct interference pattern. The experimental data and the fitted function are seen to oscillate faster than the Born results, and accordingly, c is found to be larger than $\cos\theta$ (Table I). The Born results exhibit a higher frequency at 150° than at 30°, in accordance with Eq. (3) for $q_{\mathrm{min}}\gtrsim 0$. However, the experimental data at 150° still exhibit a significantly higher frequency. This striking observation is not yet understood.

In conclusion, the present experiments confirm oscillatory structures in H_2 electron emission spectra attributed to the Young-type interferences. Furthermore, evidence is provided for the fact that the oscillation frequency of the interference pattern varies sigificantly with the electron ejection angle. Theoretical approaches provide insight into the nature of the observed interference structures. The prediction [17] that the oscillation frequency is governed mainly by the electron momentum component parallel to the beam direction is found to agree with Born calculations, and is generally confirmed by experiment, although finer details remain unexplained. In particular, neither theory explains fully the significantly higher frequency that is observed for 150°. This latter finding can be the starting point for the future work to better determine the origin of the apparently varying oscillation frequency of the oscillation pattern.

We are indebted to John Briggs, Roberto Rivarola, and Laszlo Sarkadi for very fruitful discussions. We are thankful to Ladislau Nagy for providing us with his theoretical results prior to publication. We acknowledge support from the German-French Collaboration Program PROCOPE, the financial support of the Hungarian OTKA (Grant No. T032942), the German-Hungarian S&T Collaboration (Grant No. TeT-D-17/99), the U.S. Department of Energy Office of Basic Energy Sciences, the National Science Foundation international program, and the transnational access to research infrastructures project (Grant No. HPRI-CT-1999-00019).

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