Frequency doubling of interference structures in electron emission interferences from H_2 by 68-MeV/u Kr^{33+} impact

N. Stolterfoht,[1] B. Sulik,[2] B. Skogvall,[1] J. Y. Chesnel,[3] F. Frémont,[3] D. Hennecart,[3] A. Cassimi,[3] L. Adoui,[3] S. Hossain,[4] and J. A. Tanis[4]

[1]*Hahn-Meitner-Institut Berlin GmbH, D-14109 Berlin, Germany*
[2]*Institute of Nuclear Research (ATOMKI), H-4001 Debrecen, Hungary*
[3]*Centre Interdisciplinaire de Recherche Ions Lasers, 14050 Caen cedex 04, France*
[4]*Western Michigan University, Kalamazoo, Michigan 49008, USA*

(Received 16 June 2003; published 9 January 2004)

Interference structures associated with the emission of electrons from H_2 by fast Kr^{33+} ions ($v/c=0.3$) are found to exhibit oscillations of second order superimposed on the main oscillatory structure. The secondary oscillations occur with about twice the frequency of the main oscillations. While the primary structure is produced by the coherent emission of electrons from the two atomic centers, similar to Young's two-slit experiment, our theoretical analysis indicates that the frequency doubling is a second-order effect, where the electron wave emitted at one center interferes with the wave backscattered at the other center.

DOI: 10.1103/PhysRevA.69.012701 PACS number(s): 34.50.Fa, 32.80.Fb

Single ionization in atomic collisions is a fundamental process that has attracted considerable interest for several decades since Bethe conducted his pioneering work in this field [1]. While considerable attention has been paid to the ionization of molecular hydrogen (see Refs. [2,3], and references therein), comparatively little is known about phenomena associated with coherent electron emission from the indistinguishable atomic centers. In the simplest case, ionization of H_2 resembles Young's two-slit experiment where the atomic H centers (or slits) simultaneously emit radial waves, leading to interferences in the electron emission. Such interference effects consequently reveal the wave aspect of electrons.

Early studies of collisionally induced interferences from H_2 focused on the processes of electron capture [4] and photoionization [5]. These studies were followed by additional theoretical work (see Refs. [6,7] and references therein), whereas experimental work remained limited [8]. Related effects have been observed in investigations of heavy molecules with synchrotron radiation [9,10], where one atomic center is photoionized in an inner shell followed by electron scattering at the other center. Each of these various studies can be attributed to scattering processes of first or second order as discussed in detail by Messiah [11].

Interference effects of first order were recently observed in H_2 electron emission spectra induced by fast Kr projectiles [12]. The spectra obtained at forward observation angles $\theta=20°$ and 30° exhibit oscillatory structures in good agreement with model calculations. More recent data for 3 and 5 MeV H^+ impact show similar interference effects in electron emission from H_2 [13]. It has been recognized that dipole transitions and binary encounter processes play fundamentally different roles in the ionization process leading to interferences [12,14]. Interference effects are favored by dipole transitions which are dominant at forward angles, especially for fast projectiles. Near $\theta=90°$, binary encounter collisions are strongly enhanced [15] and, hence, first-order interferences are expected to diminish in importance [12,16].

Recent theoretical studies [14,17–19] revealed additional properties of the interference effects in H_2. Calculations [18] based on the semiclassical approximation showed explicitly that the frequency of the oscillation decreases as the emission angle increases up to 90°. Moreover, the frequency was found to depend mainly on the momentum component of the ejected electron parallel to the beam direction. This prediction was essentially confirmed by further measurements of electron emission from H_2 by fast Kr impact [16], where good agreement was also found with calculations using the Born approximation. However, various specific structures in the spectra remained unexplained.

In the present work, evidence is found for interferences of second order in collisions of 68-MeV/u Kr^{33+} with H_2. These second-order interferences occur when the electron wave emitted at one center interferes with this same wave after it is backscattered at the other center. The experimental results show an interference pattern, superimposed on the main interference structure, with an oscillation frequency about double that of the first-order oscillation. The observed doubling of the oscillation frequency is supported by model calculations obtained with the Born approximation and methods known from wave optics.

In the Born approximation the cross section for ion induced electron emission from H_2 is proportional to the square of the transition matrix element (atomic units are used throughout if not otherwise stated)

$$\frac{d\sigma_{H_2}}{d\mathbf{q}\,d\Omega\,d\epsilon}\sim|\langle\varphi_{\mathbf{k}}|e^{i\mathbf{q}\cdot\mathbf{r}}|\varphi_0\rangle|^2, \tag{1}$$

where $\mathbf{r}$ is the electron coordinate and $\mathbf{q}$ is the momentum transferred in the collision. The solid angle $d\Omega$ and the energy $d\varepsilon$ refer to the outgoing electron. The initial wave function φ_0 represents the electronic two-center state of the H_2 molecule and the final wave function $\varphi_{\mathbf{k}}$ describes the outgoing electron of momentum $\mathbf{k}$. When the initial H_2 state is approximated by the (normalized) linear combination φ_0

 ©2004 The American Physical Society

$=[\varphi_{1s}(\mathbf{r})+\varphi_{1s}(\mathbf{r}-\mathbf{d})]/N$ of atomic $1s$ states separated by the internuclear distance $\mathbf{d}$, it can be readily shown that [12,17,14]

$$\frac{d\sigma_{H_2}}{d\mathbf{q}d\Omega d\varepsilon}=\frac{d\sigma_{2H}}{d\mathbf{q}d\Omega d\varepsilon}\{1+\cos[(\mathbf{k}-\mathbf{q})\cdot\mathbf{d}]\}. \tag{2}$$

The cross section $d\sigma_{2H}/d\mathbf{q}\,d\Omega d\varepsilon$ describes electron emission from the two H atoms acting as independent particles (denoted by 2H). The term in parenthesis represents the interference caused by the *two* H centers.

To compare the theoretical results with the measured cross sections it is necessary to average over the orientation of the H_2 molecule. This average is performed in closed form yielding

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

The remaining oscillatory term shows that the averaging procedure preserves the interference features of the electron emission spectra.

For comparison with experiment, a further integration is required with respect to the momentum transfer $\mathbf{q}$. Following the work of Bethe [1], the cross section given by Eq. (3) can be separated into dipole and binary encounter terms. These terms can then be integrated by means of "peaking" approximations [12] by setting

$$\mathbf{q}\approx\mathbf{0}\quad\text{for dipole transitions,}$$

$$\mathbf{q}=\mathbf{k}-\mathbf{p}_i\approx\mathbf{k}\quad\text{for binary encounters,} \tag{4}$$

where $\mathbf{p}_i$ is the initial momentum of the bound electron. Hence, the binary-encounter process leads to a rather constant term, so that the oscillatory structure in the cross section is associated primarily with the dipole term. The further analysis by Nagy *et al.* [18] noted above, in which Eq. (3) was integrated over the momentum transfer $\mathbf{q}$, showed that the interference term is governed primarily by the electron momentum component parallel to the beam direction, i.e., $k_{\parallel}=k\cos\theta$.

The data acquisition and analysis have been described in our previous work [12,16]. The experiments were performed at the Grand Accélérateur National d'Ions Lourds (GANIL) Caen, France. 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 a 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 up to a few hundred electron volts and for several electron emission angles.

The measured cross sections are found to vary by several orders of magnitude with the electron energy [2,3]. Since the variation due to the interference term is expected to be less than a factor of 2 [see Eq. (3)], this strongly varying energy dependence must be removed in order to examine the interference structures. This was done by dividing the measured cross sections by the corresponding calculated cross sections

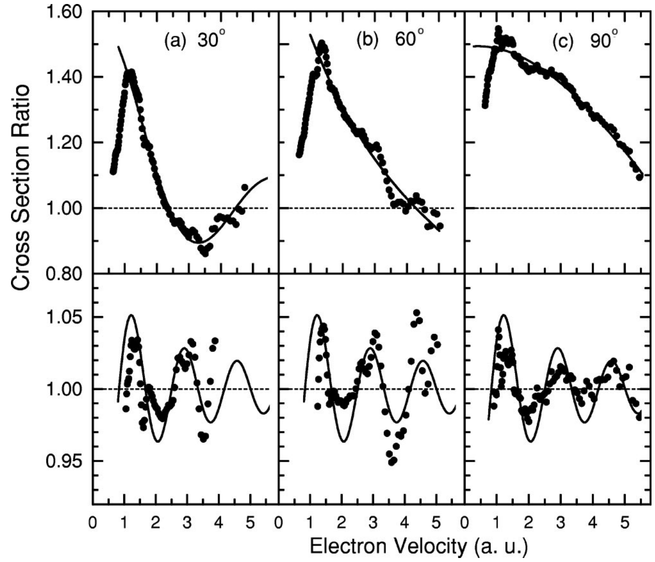

FIG. 1. Ratios of experimental to theoretical CDW-EIS cross sections for electron emission by 68-MeV/u Kr^{33+} impact on H_2 for different electron observation angles. In the upper diagrams the ratios are shown together with fits to an analytic function (see text). The ratios are divided by the corresponding fit functions and plotted in the lower diagrams together with a second-order fit.

using the continuum-distorted wave–eikonal initial state (CDW-EIS) method [20] for electron emission by two independent H atoms as described in more detail in Ref. [16]. The results for the electron observation angles 30°, 60°, and 90° are shown in the upper part of Fig. 1. These cross-section ratios are plotted versus velocity v, since the interference term is governed by the momentum k (or velocity v) of the ejected electrons [see Eq. (3)].

The cross-section ratio exhibits oscillatory structures (e.g., at 30°) that are well outside the experimental uncertainties of the relative cross sections, which are better than $\pm5\%$. Due to spurious instrumental effects larger uncertainties exist for energies below about $v\approx0.6$ a.u. (i.e., 5 eV) so that no data are shown below this value. For 30° the statistical error, increasing with velocity, becomes larger than 5% above ~4 a.u. The observed oscillatory structure has been attributed to first-order interference effects [12]. The experimental results indicate that the frequency of the oscillation varies with the electron emission angle [16] in consistency with the prediction by Nagy *et al.* [18].

To obtain information about additional spectral structures, the cross-section ratios were fit by an oscillatory function, similar to expression (3), with a variable frequency [16]

$$f(k)=F\left[1+\frac{\sin(kc\,d)}{kc\,d}\right]+G, \tag{5}$$

where F and G (with $F+G\approx1$) are the interfering and noninterfering contributions to the normalized cross section, respectively, and c is an adjustable frequency parameter. The results are shown as the solid lines in the upper diagrams of Fig. 1, and are seen to agree well with the overall structure of the data for each angle.

Careful inspection of the cross-section ratios plotted in the upper part of Fig. 1 shows, however, evidence for higher-frequency oscillations superimposed on the main oscillatory structure for each of the angles displayed. Indeed, by dividing the cross-section ratios by the fit curves just described, these secondary oscillations are clearly revealed as seen in the lower part of Fig. 1. In addition to a higher frequency, these secondary oscillations appear to have nearly equal frequencies for the electron emission angles considered here.

An attempt was made to reproduce the secondary oscillations by using a fit function similar to Eq. (5). To allow for a phase shift in the oscillation, the quantity $kc\,d$ was replaced by $kc\,d+\phi$, where ϕ is the phase shift. The resulting fit curves are given as the solid lines in the lower diagrams of Fig. 1. The fit parameters were set to have the same values for all angles, yielding $c=2.5$ and $\phi=\pi$ with fitting uncertainties of about $\pm 15\%$.

To interpret the present observations, the interference patterns are deduced from phase differences using methods known from wave optics. First, it is shown that Eq. (2) can be recovered from this method, and then the second-order contribution is analyzed. The important aspect of the present analysis is that we interpret the Born operator $e^{\mathbf{iq}\cdot\mathbf{r}}$ in Eq. (1) as an electromagnetic wave interacting with the two H centers. Thus, as shown in the upper diagram of Fig. 2, the centers labeled a and b each emit outgoing waves of momentum $\mathbf{k}$ associated with the (first-order) amplitudes A_a and A_b, respectively.

From wave optics it follows that the intensity at large distances is equal to the coherent sum $I_1=|A_a+A_b|^2$. For identical centers $|A|=|A_a|=|A_b|$ we obtain $I_1=2|A|^2(1+\cos\delta)$, where δ is the relative phase between the amplitudes. Now, $\delta=\delta_k-\delta_q$, where δ_q and δ_k are (additional) phases created along the paths crossing the centers a and b, respectively (Fig. 2). It is readily shown that $\delta_q=q\,d\cos\alpha_q=\mathbf{q}\cdot\mathbf{d}$ and $\delta_k=kd\cos\alpha_k=\mathbf{k}\cdot\mathbf{d}$ so that

$$I_1=2|A|^2\{1+\cos[(\mathbf{k}-\mathbf{q})\cdot\mathbf{d}]\}. \tag{6}$$

Consequently, by setting $2|A|^2=d\sigma_{2H}/d\mathbf{q}d\Omega\ d\epsilon$ we recover Eq. (2).

Next, we consider the interference of the first- and second-order amplitudes, which are shown in the lower diagram of Fig. 2. The diagram is restricted to one branch directed at center a, which shall be treated first. The incident wave of momentum $\mathbf{q}$ emits at center a the first-order wave with amplitude A_a which is backscattered at center b to produce a second-order wave of amplitude B_a and the outgoing momentum $\mathbf{k}$. In the latter case, the phase $\delta_d=k\,d$ is acquired as the wave propagates from one center to the other and the interference is obtained from the phase difference $\delta=\delta_k-\delta_d$. The intensity in second order restricted to the branch a is given by $I_2^a=|A_a+B_a|^2$, which yields the expression

$$I_2^a=|A_a|^2+|B_a|^2+2|A_aB_a|\cos(\mathbf{k}\cdot\mathbf{d}-k\,d). \tag{7}$$

An additional analysis, performed in analogy with the second-order treatment by Messiah [11], revealed the proportionalities for the amplitudes $A_a\sim\langle\varphi_{\mathbf{k}}|e^{\mathbf{iq}\cdot\mathbf{r}}|\varphi_{1s}\rangle$ and $B_a\sim\langle\varphi_{\mathbf{k}}|V_H|\varphi_{\mathbf{k}_d}\rangle\langle\varphi_{\mathbf{k}_d}|e^{\mathbf{iq}\cdot\mathbf{r}}|\varphi_{1s}\rangle$, where $\varphi_{\mathbf{k}_d}$ is a Coulomb wave propagating along the internuclear vector $\mathbf{d}$ and V_H is the (screened) Coulomb potential representing elastic electron scattering at a single H center.

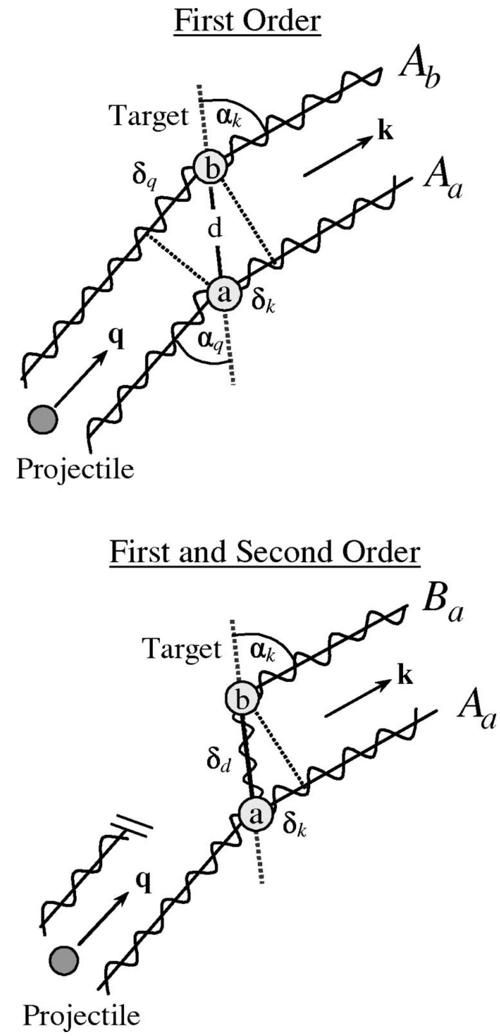

FIG. 2. Wave diagrams to visualize phase differences relevant for interferences in first and second order.

The latter expression indicates that the backscattering amplitude B_a depends on the alignment of the H_2 molecule. Thus, the average of I_2^a over the orientation of the molecule cannot be carried out in closed form, as was done to arrive at Eq. (3). Nevertheless, to obtain a qualitative understanding of the oscillation frequency produced in second order, we neglect this dependence and focus our attention instead on the phases involved in the cosine function of Eq. (7). The average can then be performed in closed form by integration over the orientation of $\mathbf{d}$ and division by 4π yielding the result

$$\overline{I_2^a}=|A_a|^2+|B_a|^2+2|A_aB_a|\frac{\sin(2k\,d)}{2k\,d}. \tag{8}$$

It is seen that the frequency of the oscillation is doubled when the primary wave is backscattered. Also, the doubling appears to be independent of the electron observation angle. These results essentially agree with our experimental observations (Fig. 1).

When the primary emission from the second center b is included, two additional waves with amplitudes A_b and B_b are created and the intensity in second order is deduced from $I_2 = |A_a + A_b + B_a + B_b|^2$. After integration over the molecular orientation (neglecting again the angular dependencies of B_a and B_b) this gives

$$\overline{I_2} = \overline{I_1} + |B_a + B_b|^2 + 2|A|(|B_a| + |B_b|) \times \left[\frac{\sin(2k\,d)}{kd} + 2\frac{\cos(k\,d)\sin(q\,d)}{qd} \right], \tag{9}$$

where $\overline{I_1}$ is the first-order interference given by Eq. (3).

Thus, the sum of the terms inside the square brackets appears as an oscillatory structure superimposed on the first-order structure described by $\overline{I_1}$. The first term in parenthesis indicates that the frequency of the second-order structure is doubled in comparison with the results of Eq. (3). The second term is more complicated. For forward angles, dipole transitions with $q \approx 0$ dominate so this latter term has the same frequency as the first-order term, however, the sine function is replaced by a cosine. For binary encounter collisions with $q \approx k$, the second term is also doubled in the oscillation frequency and adds directly to the first term.

Hence, the most pronounced oscillation of the double frequency is likely to occur at 90° where electron emission by binary encounter processes dominates. Indeed, from Fig. 1 the second-order oscillations are most clearly revealed for 90°. The same is true for 60° where binary-encounter collisions retain importance. However, also at smaller angles, e.g., 30°, we expect a secondary oscillatory contribution with a frequency doubling from the first term in parenthesis, and this result is also seen in the data of Fig. 1.

It is recalled that the frequency doubling involves a phase shift by about π in Eq. (5). This finding shows that further work is needed to examine additional phase shifts produced during the primary emission and secondary scattering at the centers. Moreover, the angular dependence of the backscattering amplitudes has to be considered to allow for an adequate integration of the second-order expressions over the H_2 orientation.

In conclusion, interference effects originating from second-order scattering at the two atomic centers of H_2 are observed. The frequency of the second-order interference pattern is predicted to be larger by a factor of 2 than the frequency obtained in first order for forward angles. This prediction is confirmed by the experimental results acquired at various electron observation angles. Second-order effects are most clearly seen at electron observation angles near 90°, where binary-encounter collisions dominate the electron emission and first-order interferences are small. Hence, evidence is provided that first- and second-order interference patterns can be observed at different observation angles in the spectra of electrons ejected from H_2 by fast projectiles.

We are indebted to Uwe Becker and Ladislau Nagy for fruitful communications. We acknowledge support from the German-French Collaboration Program PROCOPE, the Hungarian OTKA-Grant (Grant No. T032942), the German-Hungarian S&T Collaboration (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).

[1] H.A. Bethe, Ann. Phys. (Leipzig) **5**, 325 (1930).
[2] N. Stolterfoht, R.D. Dubois, and R.D. Rivarola, *Electron Emission in Heavy Ion-Atom Collisions* (Springer, Heidelberg, 1997).
[3] M.E. Rudd, Y.K. Kim, D.H. Madison, and T.J. Gay, Rev. Mod. Phys. **64**, 441 (1992).
[4] T.F. Tuan and E. Gerjuoy, Phys. Rev. **117**, 756 (1960).
[5] H.D. Cohen and U. Fano, Phys. Rev. **150**, 30 (1966).
[6] S.E. Corchs, H.F. Busnengo, and R.D. Rivarola, Nucl. Instrum. Methods Phys. Res. B **149**, 247 (1999).
[7] M. Walter and J.S. Briggs, J. Phys. B **32**, 2487 (1999).
[8] S. Cheng *et al.*, Phys. Rev. A **47**, 3923 (1993).
[9] F. Heiser *et al.*, Phys. Rev. Lett. **79**, 2435 (1997).
[10] A. Landers *et al.*, Phys. Rev. Lett. **87**, 013002 (2001).
[11] A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1970), Vol. II, p. 848.
[12] N. Stolterfoht *et al.*, Phys. Rev. Lett. **87**, 023201 (2001).
[13] S. Hossain *et al.*, Nucl. Instrum. Methods Phys. Res. A **205**, 484 (2003).
[14] M.E. Galassi, R.D. Rivarola, P.D. Fainstein, and N. Stolterfoht, Phys. Rev. A **66**, 052705 (2002).
[15] N. Stolterfoht *et al.*, Phys. Rev. Lett. **80**, 4649 (1998).
[16] N. Stolterfoht *et al.*, Phys. Rev. A **67**, 030702 (2003).
[17] J.S. Briggs, W. Ihra, and N. Stolterfoht, in *Proceedings of the XXIInd International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC)*, edited by S. Datz *et al.* (Rinton Press, Princeton, NJ, 2001), p. 565.
[18] L. Nagy, L. Kocbach, K. Pora, and J. Hansen, J. Phys. B **35**, L453 (2002).
[19] L. Sarkadi, J. Phys. B **36**, 2153 (2003).
[20] L. Gulyás, P.D. Fainstein, and A. Salin, J. Phys. B **28**, 245 (1995).