Evidence for Interference Effects in Electron Emission from H_2 Colliding with 60 MeV/u Kr^{34+} Ions

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Cross sections for electron emission in the energy range from 2–300 eV were measured for 60 MeV/u Kr^{34+} ions impacting on H_2. Model calculations are introduced to guide the search for interference effects in the electron spectra produced by the coherent emission of electrons from the two H atoms in analogy with Young's two-slit experiment. Experimentally, a full sinusoidal-like oscillation was observed in the energy range up to 250 eV in good agreement with the calculations. The oscillatory structure is found to be similar for the observation angles 20°, 30°, 150°, and 160°.

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Ionization of a target atom by the impact of a heavy ion has been the subject of intensive experimental and theoretical studies for more than five decades [1,2]. This elaborate work has been motivated by the importance of the ionization process for basic research in collision physics and for various applications in adjacent fields. Within the field of particle-induced ionization, particular attention has been devoted to the molecular target H_2 [3–6], which is the simplest molecule composed of two atoms. Since these atoms are indistinguishable, their contributions to ionization add coherently and interference effects might be expected in the ionization process. Such electron emission from H_2 may be closely related to Young's two-slit experiment, which played an important role in the pioneering period of quantum mechanics.

Studies of collisionally induced interference effects from H_2 have focused on the process of single electron capture. Early work by Tuan and Gerjuoy [7] has been followed up by various theoretical studies; see, for example, Ref. [8] and references therein. In particular, it has been shown that interference structures are preserved when the capture cross section is averaged over the orientation of the internuclear axis of the H_2 molecule [9]. Similar results have been obtained with respect to the stopping power of H_2 moving in condensed matter [10].

Interference effects in the photoionization of H_2 have been considered for many years [11–13]. Experiments with synchrotron radiation [14,15] have focused on heavier molecules where one atomic center is photoionized (e.g., in an inner shell) followed by electron scattering at the other center. In solid-state physics a method based on similar scattering effects is known as EXAFS (extended x-ray absorption fine structure) [16]. These scattering phenomena differ, however, from Young's experiment, where both slits simultaneously emit radial waves giving rise to a diffraction pattern. (An overview for both cases is given by Messiah [17].) Although much experimental work has been devoted to the ionization of H_2 by heavy particles [3–6], interference effects have not been observed in the corresponding emission spectra.

In the present Letter, we provide evidence for interferences in electron emission from H_2 using 60 MeV/u Kr^{34+} ions. We first perform model calculations to determine the spectral region where interferences can be expected. It is shown that the high projectile velocity of about 50 a.u. is advantageous as it enhances the visibility of interference effects. Then, the experimental method is presented and the measured electron spectra are found to exhibit oscillatory structures in agreement with the model predictions.

To describe interference effects for single ionization we apply a formalism analogous to that developed for electron scattering [17]. (Atomic units are used throughout if not otherwise stated.) The cross section for electron emission from the H_2 molecule can be written as

$$\frac{d\sigma_{\mathrm{H}_2}}{d\mathbf{q}\,d\Omega\,d\epsilon} = \frac{d\sigma_{2\mathrm{H}}}{d\mathbf{q}\,d\Omega\,d\epsilon}[1 + \cos(\mathbf{p}\cdot\mathbf{d})], \qquad (1)$$

where the solid angle $d\Omega$ and the energy $d\epsilon$ refer to the outgoing electron. The cross section $d\sigma_{2\mathrm{H}}/d\mathbf{q}\,d\Omega\,d\epsilon$ describes electron emission from the two H atoms acting as independent particles (denoted by the label 2H). The term in parentheses represents the interference caused by the *two* H centers where $\mathbf{d}$ is the vector associated with the internuclear distance of the H_2 molecule and $\mathbf{p} = \mathbf{k} - \mathbf{q}$ is the difference between the outgoing electron momentum $\mathbf{k}$ and the momentum transfer $\mathbf{q}$. A similar formula can be deduced for electron emission from two slits, when the momentum $\mathbf{p}$ is replaced by $\mathbf{k}$ in Eq. (1).

Since the recoil ions are not observed in this work, we perform an averaging (integration and division by 4π) over the orientation of the internuclear axis of H_2, which can be done analytically [9,10]

$$\frac{d\sigma_{H_2}}{d\mathbf{q}\,d\Omega\,d\epsilon} = \frac{d\sigma_{2H}}{d\mathbf{q}\,d\Omega\,d\epsilon}\left[1 + \frac{\sin(pd)}{pd}\right]. \quad (2)$$

Hence, the averaging procedure preserves the essential features of the interference in the electron emission spectra. However, it cancels the dependence of the interference term on the electron emission angle θ.

To obtain the double differential cross sections relevant for the present experiment, an integration over $\mathbf{q}$ is necessary. Following the pioneering work by Bethe [18] the cross section is split into a dipole part representing soft collisions and a binary part representing violent collisions between the projectile and the target electron. The dipole part has a sharp maximum at the minimum momentum transfer $q_{\min} = \Delta E/v_p$ where ΔE is the energy transfer and v_p is the projectile velocity. For fast projectiles ($v_p \approx 50$ a.u.) $q_{\min}$ is small, so we can apply a peaking approximation, where q is set to zero in the interference term and, thus, $p = k$ as in the two-slit experiment. A similar peaking approximation can be performed for the binary part by setting p equal to the mean initial momentum p_i of the bound electron [19]. Hence, we obtain the integrated cross section

$$\frac{d\sigma_{H_2}}{d\Omega\,d\epsilon} = \frac{d\sigma_{2H}^{\text{dip}}}{d\Omega\,d\epsilon}\left[1 + \frac{\sin(kd)}{kd}\right] + \frac{d\sigma_{2H}^{\text{bin}}}{d\Omega\,d\epsilon}\,s\,, \quad (3)$$

where $d\sigma_{2H}^{\text{dip}}/d\Omega\,d\epsilon$ and $d\sigma_{2H}^{\text{bin}}/d\Omega\,d\epsilon$ are integrated cross sections referring to the dipole and binary part, respectively, and $s = 1 + \sin(p_i d)/(p_i d)$ with $p_i \approx 1$ a.u. is a constant. It is important to note that with constant s only the dipole term is responsible for an oscillation in the cross section. Therefore, the high projectile velocity is important, since it enhances the dipole term [19].

Unlike the interference term, the cross section associated with the independent H atoms is strongly dependent on the electron emission energy and angle. These cross sections may vary with energy by several orders of magnitude [1,2], whereas the variation due to interference effects is limited to a factor of 2 as can be seen from Eq. (3). Therefore, to observe the interference effects, it is necessary to remove the strong energy and angle variations of the cross sections. This is done by normalizing (dividing) the cross section for H_2 by $d\sigma_{2H}/d\Omega\,d\epsilon$:

$$\left(\frac{d\sigma_{H_2}}{d\Omega\,d\epsilon}\right)_{\text{nor}} = D\left[1 + \frac{\sin(kd)}{kd}\right] + Bs \quad (4)$$

where the subscript nor denotes the cross section normalization. We use D and B (with $D + B = 1$) as abbreviations for, respectively, the normalized dipole and binary cross sections, which are slowly varying with k.

Returning to the interference effects we note that Eq. (4) predicts a full sinusoidal oscillation of the normalized cross section when the product kd varies from 0 to 2π. Since the internuclear separation $d = 1.42$ a.u. for H_2, this oscillation is governed by k varying from 0–4.3 a.u. Recalling that $k = \sqrt{2\varepsilon}$ it follows that an oscillatory structure should be expected in the range of electron energies $\varepsilon = 0$–250 eV.

The experiments were carried out at the GANIL accelerator facility in Caen, France. The scattering chamber and the electron spectrometer were similar to those used previously [19,20] so that only a brief description shall be given here. A beam of 60 MeV/u Kr^{34+} ions with a current of 1–2 μA was collimated to a size of about 2 mm $\times$ 2 mm. It was directed onto an H_2 target of $\sim$4 mm diameter obtained by means of a gas jet. Continuum electrons emitted from the target were measured with a parallel-plate electron spectrometer for energies ranging from about 2–300 eV, and for angles of 20°, 30°, 150°, and 160°. Absolute cross sections for electron emission were determined by methods similar to those used in previous work [20]. Auxiliary measurements were performed with He to verify the reliability of the H_2 results. Earlier He ionization measurements with fast projectiles [20,21] have shown that the experimental data are well reproduced by theoretical calculations [22].

Typical results for electron emission at 30° and 150° from He and H_2 are shown in Figs. 1(a) and 1(b), respectively. Also plotted are theoretical cross sections obtained

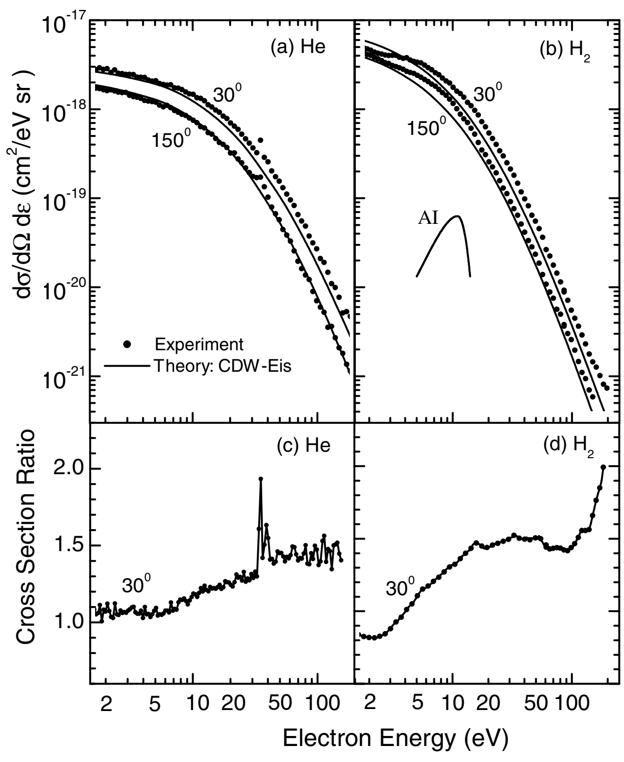

FIG. 1. Cross sections and ratios for electron emission by 60 MeV/u Kr^{34+} impacting on He and H_2 as a function of the ejected electron energy. In (a) and (b) experimental and theoretical cross sections are compared for He and H_2, respectively, obtained at the observation angles 30° and 150°. In (b) the curve labeled "AI" is due to autoionization (see text). In (c) and (d) cross section ratios of experimental and theoretical results are given for He and H_2, respectively, obtained at 30°.

by means of the continuum-distorted-wave eikonal-initial-state approximation (CDW-EIS) assuming independent H atoms. The calculations for H were performed using hydrogenic wave functions with an effective target charge of $Z_t = 1.05$ [23], and the He data were calculated using Hartree-Slater wave functions for the initial and final electron states [22]. The cross sections are seen to vary strongly, as mentioned before.

To enhance the visibility of possible interference structures, the measured cross sections were divided by the corresponding theoretical cross sections. The results for 30° associated with He and H_2 are given in Figs. 1(c) and 1(d), respectively. The He data show a distinct peak near 33 eV produced by autoionization [20]. Apart from this peak structure, the He data show a smooth monotonic increase with increasing energy of the ejected electrons. At low energies the cross section ratio is close to unity, whereas at higher energies it deviates from unity indicating an increasing disagreement between experiment and the CDW-EIS model [20,21].

On the other hand, the cross section ratio for H_2 indicates a nonmonotonic increase suggesting an oscillatory structure. This structure is well outside the experimental uncertainties of the relative cross sections which are typically $\pm 10\%$. Larger uncertainties are expected at energies smaller than ~ 5 eV where the measured cross sections can be affected by spurious instrumental effects. However, from the low-energy data of He [Fig. 1(c)] it may be inferred that the experimental setup is capable of reliably measuring electrons at energies as low as 2 eV. At high energies, i.e., above about 150 eV, the measured cross sections become less accurate, since they are increasingly influenced by limited statistics and an underlying background. At energies of about 250 eV the uncertainties of the cross sections for the forward angles are estimated to be as high as $\pm 25\%$.

An important consideration for the H_2 spectra is the possible contribution of autoionization electrons. A doubly excited H_2 molecule dissociates, sharing its energy between the recoiling fragments and autoionization electron [24]. The energy distribution of these autoionization electrons was estimated using recoil energy data from H_2 fragmentation experiments [25]. This distribution can be put on an absolute scale using the double excitation cross section of $\sim 4 \times 10^{-19}$ cm^2/sr obtained by extrapolating previous data [25,26] and assuming isotropic electron emission. The resulting cross sections are given by the curve labeled "AI" in Fig. 1(b). [We note that this cross section for H_2 is consistent with the corresponding values of $(2-3) \times 10^{-19}$ cm^2/sr derived from the present He autoionization lines shown in Fig. 1(a).] Integration of the measured H_2 spectra in Fig. 1(b) within the range of 5–15 eV, where H_2 autoionization occurs, yields cross sections $\sim 2 \times 10^{-17}$ cm^2/sr which are a factor of 50 larger than the H_2 autoionization values. Thus, we can conclude that autoionization contributes negligibly to the present H_2 spectra.

We recall that Eq. (4) predicts a sinusoidal structure as a function of the momentum k (or velocity v). Accordingly, we plot the present cross section ratios versus the velocity v in Fig. 2, where results for the electron emission angles of 20°, 30°, 150°, and 160° are given. To show the overall increase of the cross section ratio with velocity, we fit straight lines to the data indicated by the dashed curve representing the linear function $c(v) = a + bv$. Similar to the case of He, this increase can partially be attributed to the discrepancies between experiment and the CDW-EIS model. The dashed lines enhance the visibility of the spectral structures. Indeed, sinusoidal-like oscillations are clearly seen at forward angles and, to a lesser extent, also at backward angles. However, we expect certain differences between forward and backward angles due to higher-order scattering effects as observed in photoionization studies [14,15].

To allow for a direct comparison with theory, the straight line fit was used to remove the overall increase of the cross section ratios. The ratios divided by the $c(v)$ function are given in Fig. 3 for the angles of 20° and 30°. In this plot, the sinusoidal oscillations become even more visible. The figure also shows model calculations from Eq. (4) using the estimated values $D = 0.65$ and $B = 0.35$ (see the formula for $d\sigma_{2\mathrm{H}}/d\mathbf{q}\, d\Omega\, d\epsilon$ in, e.g., Ref. [2]). The actual values are of minor importance, since we are primarily interested in the velocity dependence. The good agreement

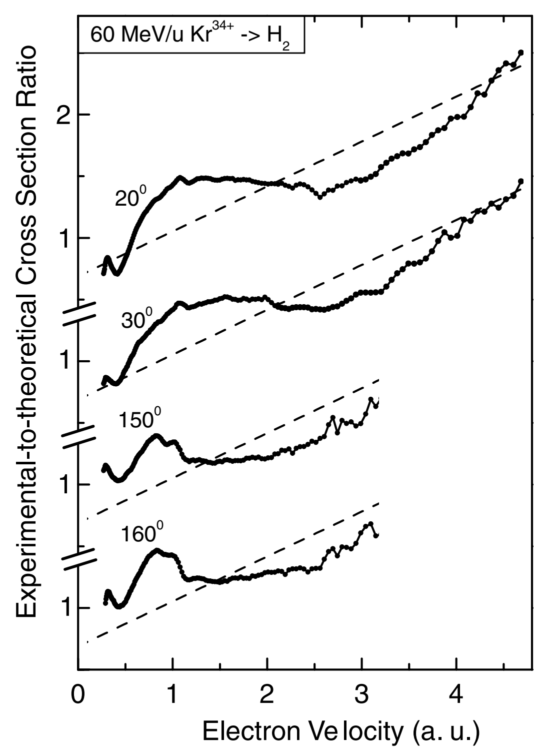

FIG. 2. Experimental-to-theoretical cross section ratios for the collision system 60 MeV/u Kr^{34+} + H_2. The electron observation angles are 20°, 30°, 150°, and 160°. The dashed lines are obtained from a linear function introduced to fit the overall increase of the cross section ratios.

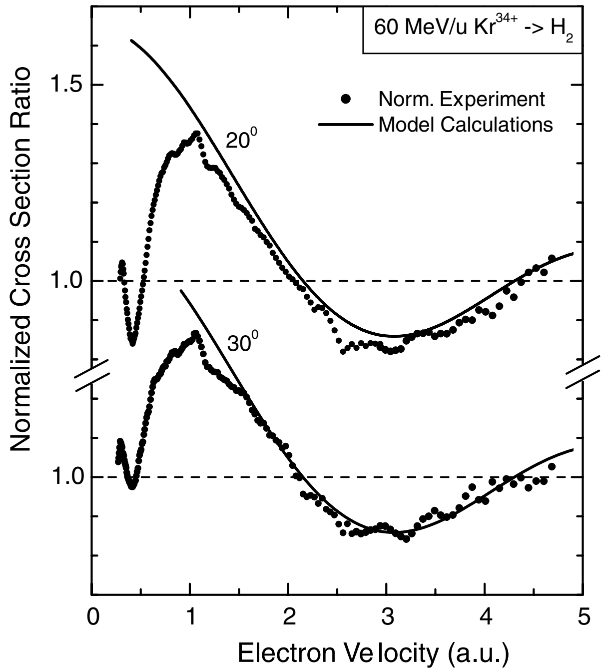

FIG. 3. Experimental-to-theoretical cross sections divided by the linear fit function shown as dashed lines in Fig. 2. The electron observation angles are 20° and 30° as indicated. The solid lines represent model calculations from Eq. (4).

found between the calculations and the normalized experimental data provides strong evidence that the structures in the electron spectra can be associated with interference effects caused by the two atomic centers of H_2. In particular, we observe one full oscillation within the velocity range 0–4.3 a.u. in accordance with the theoretical predictions.

Because of the instrumental difficulties of measuring very slow electrons, the cross sections below ~0.5 a.u. are uncertain. However, since the experimental data are expected to be reliable for velocities greater than 0.5 a.u., the model results should be regarded with caution in the velocity range of about 0.5–1 a.u. Specifically, at small velocities the peaking approximations applied in the present model are not expected to be valid. Moreover, near 1 a.u. the measured cross section ratios exhibit certain reproducible wiggles which may indicate higher-order effects in the electron scattering at the two H centers [17].

In conclusion, we have observed structures attributed to interference effects in the electron emission spectra from H_2, which are similar to those observed in Young's two-slit experiment. For ion-induced electron emission the interference structures are difficult to observe. Therefore we have used theory to provide guidance in the search for the interference phenomena. We found that fast projectiles are essential, since they enhance dipolelike transitions which, in turn, are responsible for the interference effects. Various features predicted by the present model calculations are in accordance with the experimental results: (i) interference effects do not cancel when an averaging is performed over the orientation of the H_2 internuclear axis, (ii) interference effects are manifested by a sinusoidal-like oscillation occurring in an energy range up to 250 eV, and (iii) the oscillatory structures are rather independent of the observation angles. Further work is planned to improve the statistics for fast electrons to extend the observation range of the sinusoidal oscillation. Moreover, additional effort is required to verify spectral structures that may be due to interference effects of higher order.

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