

Interference in the Collective Electron Momentum in Double Photoionization of H₂

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We investigate single-photon double ionization of H₂ by 130 to 240 eV circularly polarized photons. We find a double slitlike interference pattern in the sum momentum of both electrons in the molecular frame which survives integration over all other degrees of freedom. The difference momentum and the individual electron momentum distributions do not show such a robust interference pattern. We show that this interference results from a non-Heitler-London fraction of the H₂ ground state where both electrons are at the same atomic center.

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Photoelectron emission from H₂ closely resembles the situation of an electron wave downstream from a double slit. This was first realized by Cohen and Fano in 1966 [1] and Kaplan and Markin [2]. They predicted that photoelectron emission from diatomic molecules will exhibit a two center interference pattern in the angular distribution of the electron with respect to the molecular axis.

For single-electron emission the situation is well studied theoretically (see, e.g., [2–7]) and experimentally demonstrated for ion [8,9] and photon impact [10,11]. The case of emission of two electrons, corresponding to double ionization, however, is much less clear. Electron correlation is responsible for single-photon double ionization and the corresponding experiments probe the very small part of the wave function that exhibits this electron correlation. The key question here is what is the interplay between electron correlation and single particle interference. Does electron-electron correlation destroy the coherence of the two centers? Or alternatively how does the two-center nature change the electron correlation pattern? One of the unique and fundamental aspects of the H₂ double slit experiments is the ability to measure the momentum imparted to the “slit.” In this Letter we will show that the recoil momentum of the slit exhibits very strong interference fringes. The recoil of the slit balances the sum momentum of the two outgoing electrons. Using simple plane wave arguments without entering into the details of electron correlation we find that the part of the wave function responsible for double ionization has the two electrons originating from the same center at one hydrogen or the other.

For the description of two electron processes two alternative coordinate frames have proven useful: The individual electron momenta k_1 and k_2 and the corresponding

Jacobi coordinates $k^+ = k_1 + k_2$ and $k^- = 1/2(k_1 - k_2)$. k^+ corresponds to the center of mass motion of the electron pair, which by momentum conservation is the recoil momentum imparted onto the two protons in H₂ [12–14].

Walter and Briggs have argued on theoretical grounds [3] that the two center interference pattern will be visible in the angular distribution of the Jacobi momentum k^- even if k^+ is unobserved. On the contrary, Berakdar *et al.* have found theoretically [15] that, for the related situation of double electron ejection from a crystalline surface, an interference structure is visible in k^+ . Finally we have recently shown that in the angular distribution of individual electron momenta $k_{1,2}$ the Cohen Fano type interference is visible only for extremely unequal energy sharing [16]. If, however, the second electron (k_2) carries more than about 10% of the excess energy, integration over k_2 leads to decoherence and loss of the interference structure in k_1 .

In this Letter we will examine the interplay of electron correlation and two center interference in double photoionization of H₂ using Jacobi (k^+ , k^-) and single-electron (k_1) coordinates for excess energies of 80–190 eV. Such multiple coincidence experiments on H₂ are technically highly demanding and became feasible only recently [16–19]. Our data show a surprisingly robust interference pattern in k^+ . Our analysis presented below shows that this interference results from the fact that the electron pair emerges from the same proton of the H₂, which is only a small contribution to the H₂ ground-state wave function.

To allow a transformation of experimental data from individual electron momenta to Jacobi coordinates the full final state phase space of both electrons and both ions has to be measured with no preselection on angles or energies. For this we have used the COLTRIMS technique

[20–22] to image electrons and ions from double photoionization at 130, 160, 200, and 240 eV photon energy. We have crossed the photon beam from beam line 11 of the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory with a well-collimated cooled supersonic H_2 beam. The electrons and ions are collected by a 36 V/cm electric field and a superimposed homogeneous magnetic field of 14.7 G on two multichannel plate detectors with delayline anodes [23].

The four momentum vectors of the two electrons and the two protons in the final state of the reaction are interrelated by momentum conservation. Therefore only three of the fragments have to be measured directly, and the fourth one can be deduced from momentum conservation law. We have chosen to detect both protons and the lower energy electron and calculate the momentum of the faster electron. While this is a frequently used procedure in ionization processes of atomic targets [24–26], it is challenging for molecular targets which dissociate. In this case the main source of momentum on the ionic fragments of a molecule is the Coulomb repulsion of the nuclei with about 36 a.u. for H_2 while the recoil of the center of mass of the two protons from the high energy electron emission is only about 3 a.u. in the present case. Therefore a high momentum resolution on a rather large ion momentum has to be achieved in order to extract this recoil momentum from the proton momenta.

Figure 1 shows the key result of the present work. We compare the angular distribution of k_1 , k^+ , and k^- in the molecular frame. The top row shows the angular distribution of k_1 , k^+ , and k^- against the magnitude of the respective momentum. The interference is visible in the momentum k_1 of the individual electron only for extremely asymmetric energy sharing [Fig. 1(c)]. For more symmetric sharing [Fig. 1(d)] the second electron, which is integrated over in this presentation, leads to decoherence of the electron wave [16]. The k^+ coordinate, to the contrary, shows a very pronounced interference pattern, while k^- does not show any interference.

To analyze the origin of this observation we generalize the arguments of Cohen and Fano [1]. We take four possible breakup routes of the 4-body system into account, as depicted in Fig. 2. Paths *a* and *b* are both electron waves with individual momenta k_1 and k_2 emerging both from the same center either right or left, path *c* and *d* correspond to both electrons emerging from opposite centers. The experiment will observe the coherent sum of all 4 paths. In a homonuclear molecule path *a* and *b* as well as path *c* and *d* are indistinguishable so that we end up with two paths

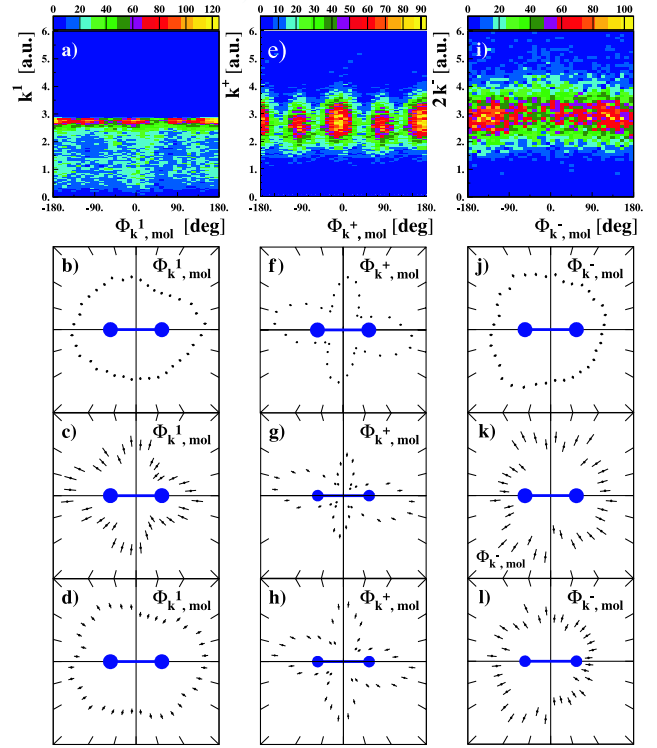


FIG. 1 (color online). Double photoionization of H_2 by 160 eV circular polarized photons. (a) Horizontal axis: angle between k_1 and the molecular axis in the polarization plane, vertical axis: $|k_1|$. k_1 and the molecular axis are each within $\pm 20^\circ$ of the polarization plane; there is no restriction on k_2 . (b) Polar presentation of data of (a) integrated over $|k_1|$. (c) Same for $2.75 \text{ a.u.} < |k_1| < 2.81 \text{ a.u.}$. (d) Same for $2 \text{ a.u.} < |k_1| < 2.7 \text{ a.u.}$ (e) Horizontal axis: angle between k^+ and the molecular axis in the polarization plane, vertical axis: $|k^+|$. k^+ and molecular axis are each within $\pm 20^\circ$ of the polarization plane; the data are integrated over all other observables. (f) Polar presentation of data of (e) integrated over $|k^+|$. (g) Same for $3.1 \text{ a.u.} < |k^+| < 3.9 \text{ a.u.}$. (h) Same for $2.3 \text{ a.u.} < |k^+| < 2.6 \text{ a.u.}$. (i) Horizontal axis: angle between k^- and the molecular axis in the polarization plane, vertical axis: $|2k^-|$. k^- and molecular axis are each within $\pm 20^\circ$ of the polarization plane; the data are integrated over all other observables. (j) Polar presentation of data of (i) integrated over $|2k^-|$. (k) Same for $3.5 \text{ a.u.} < |2k^-| < 3.8 \text{ a.u.}$ (l) Same for $2.2 \text{ a.u.} < |2k^-| < 2.5 \text{ a.u.}$

α) (both electrons from the same center) and β (both electrons from opposite centers). For simplicity we assume plane waves for the electrons, which yields to a simple product wave function. For the momentum distribution $W(\vec{k}_1, \vec{k}_2)$ one obtains:

$$\begin{aligned}
 W(\vec{k}_1, \vec{k}_2) &= |\alpha[e^{i\vec{k}_1 \cdot (\vec{r}_1 + \vec{R}/2)} e^{i\vec{k}_2 \cdot (\vec{r}_2 + \vec{R}/2)} + e^{i\vec{k}_1 \cdot (\vec{r}_1 - \vec{R}/2)} e^{i\vec{k}_2 \cdot (\vec{r}_2 - \vec{R}/2)}] + \beta[e^{i\vec{k}_1 \cdot (\vec{r}_1 + \vec{R}/2)} e^{i\vec{k}_2 \cdot (\vec{r}_2 - \vec{R}/2)} + e^{i\vec{k}_1 \cdot (\vec{r}_1 - \vec{R}/2)} e^{i\vec{k}_2 \cdot (\vec{r}_2 + \vec{R}/2)}] |^2 \\
 &= 4|\alpha \cos(\vec{k}^+ \cdot \vec{R}/2) + \beta \cos(2\vec{k}^- \cdot \vec{R}/2)|^2 \\
 &= 4\alpha^2 \cos^2(\vec{k}^+ \cdot \vec{R}/2) + 4\beta^2 \cos^2(2\vec{k}^- \cdot \vec{R}/2) + 4\alpha\beta[\cos(\vec{k}_1 \cdot \vec{R}) + \cos(\vec{k}_2 \cdot \vec{R})]
 \end{aligned} \tag{1}$$

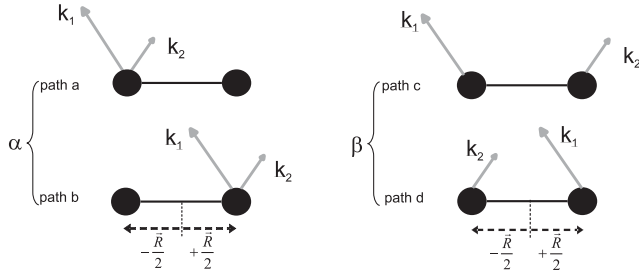


FIG. 2. Four different interfering breakup channels contributing to double electron ejection from H_2 ; see text and Eq. (1).

Here \vec{R} is the internuclear distance and the two protons are located at $-\vec{R}/2$ and $+\vec{R}/2$, \vec{r}_1 and \vec{r}_2 denote the coordinates of electrons 1 and 2. α and β are the amplitudes for the two possible pathways.

Equation (1) shows that configuration α results in an interference pattern on k^+ , while configuration β leads to an interference in k^- . If all pathways contribute one obtains an additional cross term.

Our data clearly show that configuration α by far dominates; we see no trace of the interference in k^- . That indicates clearly that the two electrons have originated from the same center and that single-photon double ionization at this photon energy proceeds mainly through this ionic contribution of the H_2 ground state. This is only known for strong field double ionization and can be attributed, in that case, to the field which first pulls both electrons to one side before they are ejected [27,28].

Our observations do not agree with the prediction by Briggs and Walter [3] which is not surprising since they use a Heitler-London wave function which is the dominant part of the H_2 ground state [29]. It has the two electrons centered at two different protons. In contrast, in Ref. [15] Berakdar *et al.* assume the electrons emerge from the same center on a crystalline surface leading to $[\cos(\vec{k}^+ \cdot \vec{R}/2)]^2$ in agreement with our measurement. Our data suggest that the photo double ionization process selectively picks this small fraction of the ground state.

We now investigate the photon energy dependence of this finding. Figure 3 shows the angular distribution for photon energies of 130, 160, 200, and 240 eV. All energies show qualitatively similar behavior for k^+ . Figure 3 shows the highest interference contrast for the highest photon energy. For all photon energies the interference contrast is highest for the largest value of k^+ . The maximum value of k^+ for a given photon energy corresponds to both electrons being emitted in parallel. Note that at all photon energies the angular distribution is rotated clockwise with respect to the molecular axis. The rotation angle decreases with photon energy. This rotation is a consequence of circular dichroism associated with circular polarization used in this experiment [30,31]. It results from multiple scattering of the wave in the molecular potential. In Eq. (1) one neglects any multiple scattering contributions. The

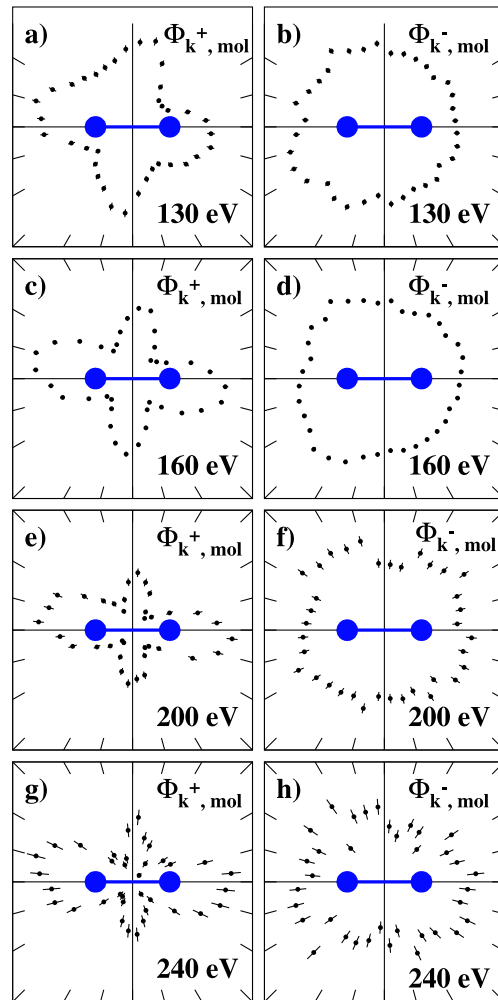


FIG. 3 (color online). Angular distribution of k^+ (left column) and k^- (right column), for different photon energies as given in the figure. Photon propagation into the plane of the figure, molecular axis, and k^+ or k^- are restricted to be within $\pm 20^\circ$ of the polarization plane. It is integrated over the magnitude of k^+ and $2k^-$ and over all other observables.

reason to use circular polarized instead of linear polarized light was to avoid a quasinode in the angular distribution at 90° to the polarization axis, which results from the polarization alone and has nothing to do with the two center nature of the problem. For circular polarization and in the polarization plane all angular distributions would be strictly isotropic for atoms and hence any structure can uniquely be attributed to molecular effects.

Our experimental observations (Fig. 1) of the interference in the center of mass momentum k^+ suggest to view double ionization as an ejection of a quasiparticle of mass 2, i.e., a dielectron. This particle is emitted from either center of the molecule. Its center of mass motion with momentum k^+ experiences the two center nature of the problem and hence exhibits the interference pattern as it is

known from single-electron emission. The fragmentation of this dielectron leads to the momentum k^- . It is in first order independent of the two center nature of the problem and it is governed by electron correlation.

Our observations are not a small feature in a restricted region of the 10 dimensional phase space of this 4-body problem. The observed interference in the electron center of mass motion is the dominant feature governing the total double ionization process. Our result shows that double ejection via the dominant Heitler-London part of the H_2 ground state does not significantly contribute to double ionization in H_2 . We conclude that double ionization proceeds via the ionic part of the H_2 ground state. Our result very clearly links the observable final state to a particular feature in the initial ground state. This connection has been searched for in double ionization processes [32,33] but has rarely been found [19,34–36].

We suggest to make use of a full calculation as in [37,38] to investigate the origin of the observed interference in full detail. Our results suggest that an inspection of the outgoing two body wave function in the near field of the molecule will show the two electrons be born close together in space near one or the other nucleus. This conclusion is also supported by recent theoretical work of Colgan *et al.* [39].

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