

## Photo-double-ionization of H<sub>2</sub>: Two-center interference and its dependence on the internuclear distance

M. S. Schöffler,<sup>1</sup> K. Kreidi,<sup>1,2</sup> D. Akoury,<sup>1,3</sup> T. Jahnke,<sup>1</sup> A. Staudte,<sup>1</sup> N. Neumann,<sup>1</sup> J. Titze,<sup>1</sup> L. Ph. H. Schmidt,<sup>1</sup> A. Czasch,<sup>1</sup> O. Jagutzki,<sup>1</sup> R. A. Costa Fraga,<sup>1</sup> R. E. Grisenti,<sup>1</sup> M. Smolarski,<sup>1</sup> P. Ranitovic,<sup>4</sup> C. L. Cocke,<sup>4</sup> T. Osipov,<sup>3</sup> H. Adaniya,<sup>3</sup> S. Lee,<sup>3</sup> J. C. Thompson,<sup>5</sup> M. H. Prior,<sup>3</sup> A. Belkacem,<sup>3</sup> Th. Weber,<sup>3</sup> A. Landers,<sup>5</sup> H. Schmidt-Böcking,<sup>1</sup> and R. Dörner<sup>1,\*</sup>

<sup>1</sup>*Institut für Kernphysik, University Frankfurt, Max-von-Laue-Strasse 1, D-60438 Frankfurt, Germany*

<sup>2</sup>*DESY, Notkestrasse 85, 22607 Hamburg, Germany*

<sup>3</sup>*Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

<sup>4</sup>*Department of Physics, Kansas State University, Cardwell Hall, Manhattan, Kansas 66506, USA*

<sup>5</sup>*Department of Physics, Auburn University, Auburn, Alabama 36849, USA*

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We investigate photo-double-ionization of H<sub>2</sub> by circular polarized photons at  $h\nu=160$  eV. The previously observed two-center interference pattern in the angular distribution of the electron sum momentum is shown to depend strongly on the internuclear distance and the summed electron momenta.

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Double ionization of He and H<sub>2</sub> by a single photon is one of the simplest test cases for few body quantum dynamics and the study of electron correlation, beside the simplest mechanism, the double excitation [1]. Tremendous progress has been made experimentally and theoretically in recent years for both systems. For helium today within the dipole approximation exact theoretical solutions are available. Good agreement is found with a rich body of experimental data from a wide energy range of 100 meV [2] to 450 eV [3,4] above the double ionization threshold (see [5,6] for reviews). Compared to He, H<sub>2</sub> adds two new degrees of freedom to the problem: the internuclear axis and the internuclear distance  $R$ . The most intriguing effect resulting from this two-center nature of the problem is a double-slit-like interference in the electron emission pattern. For single electron emission it was first noted by Cohen and Fano in 1966 [7] that the electron wave launched by photoionization of a homonuclear diatomic molecule is very similar to the wave behind a double slit. This leads to oscillation of the cross section even after integration over the orientation of the molecule [7,8]. The traces of this two-center interference in the ionization continuum of H<sub>2</sub> has been seen for electron emission by ion impact [9–15]. Most clearly, however, the interference can be seen in the electron angular distribution in the body fixed frame. This angular emission pattern is the direct analog to the fringe pattern on the screen behind the double slit. First calculations for H<sub>2</sub> in a plane wave approximation are reported by Kaplan and Markin [16]. In the plane wave approximation, one obtains an angular distribution given by

$$W(\Phi) = \cos\left(\vec{k} \cdot \frac{\vec{R}}{2}\right)^2 = \cos\left[k \frac{R}{2} \cos(\Phi)\right]^2, \quad (1)$$

where  $\Phi$  is the angle between the electron and the molecular axis,  $R$  is the internuclear distance, and  $k$  the momentum of

the electron [17]. Equation (1) is the result of the superposition of two spherical waves, i.e. it describes the optical double slit exactly. Several more refined calculations have shown that for a photoelectron wave emerging from H<sub>2</sub> the molecular potential and multiple scattering effects do not obscure this interference effect [17–20].

In our previous work [21] we have shown that an interference pattern according to Eq. (1) can be seen for double ionization only for the case of extremely unequal energy sharing. For  $E_\gamma=240$  eV and an energy of  $<5$  eV for the slow and 185–190 eV for the fast electron exhibits a clear interference pattern. For more equal energy sharing, however, the interference in the angular distribution of one electron is lost, if the second electron is not observed. It can be recovered by measuring the second electron and fixing the relative angle between the two electrons. The loss of interference by electron-electron interaction in double ionization can be overcome if instead of the angular distribution of one of the electrons, the angular distribution of the sum momentum  $k_e^+ = k_{e1} + k_{e2}$  of both electrons is measured [22]. This observable shows a very robust interference pattern which survives the integration over all other degrees of freedom.

For a simple intuitive argument why for double ionization  $k_e^+$  rather than the individual electron momenta shows the interference, one might think of the electron pair as one composite quasiparticle of momentum  $k_e^+$ , i.e. a dielectron. This quasiparticle then decays in two subparticles, the two individual electrons. Such a decay will not change the sum momentum  $k_e^+$  and hence preserve the interference pattern on this quantity. More formally, we have obtained the same result by generalizing the arguments of Cohen and Fano in [22]. One finds that the interference in  $k^+$  is the result of both electrons being ejected together from the left or the right nucleus (see Kreidi *et al.* in [22]).

The present work is the third of this series of experimental papers [21,22] in which we study the influence of the two-center interference on double-photoionization of H<sub>2</sub>. Here we show a strong dependence of the interference effect on the internuclear distance  $R$ . From the double slit analogy and Eq.

\*doerner@atom.uni-frankfurt.de

(1) it is obvious that such a dependence can be expected. It can be experimentally accessed in  $H_2$  since even in its vibrational ground state the molecule offers a range of internuclear distances (0.6–0.9 Å at FWHM). By measuring the kinetic energy release (KER) of the two protons after double ionization one can determine  $R$  at the instant of electron ejection from the measured KER using  $R=1/\text{KER}$  [23–26]. For the purpose of the present study on the internuclear distance dependence we consider the angular distribution of  $k_e^+$  instead of that of the individual electron momenta [22].

Technically, we have measured  $k_e^+$  by measuring the momenta of both protons  $k_{p1,2}$  with a very high accuracy. The sum momentum of both protons is by momentum conservation equal and opposite to the sum momentum of both electrons. The proton momenta are best described by the sum momentum  $k_p^+$  and the difference or relative momentum  $k_p^-$  between the two protons.  $k_p^-$  is generated by the repulsion. At  $R=0.75$  Å one obtains  $k_p^-=36$  a.u. It is much larger than the maximum possible  $k_p^+$ , which one obtains for the very unlikely case that two electrons emerge with parallel momenta and equal energy sharing. At the present photon energy of 160 eV the maximum  $k^+$  is 2.8 a.u. (ionization potential of  $H_2=54$  eV), while  $k_p^-$  resulting from the equilibrium distance of 0.75 Å is 36 a.u. corresponding to a KER of 19.2 eV.

The experiment has been performed at beamline 11.0.2 of the Advanced Light Source at Lawrence Berkeley National Laboratory using the COLd target recoil ion momentum spectroscopy (COLTRIMS) technique [27,28]. The experimental setup is very similar to the one described in [29]. In brief, the focused circular polarized photon beam is crossed with a supersonic beam of  $H_2$ . Electrons and ions created in the interaction volume are guided by an electrostatic field of 36 V/cm and a parallel homogenous magnetic field [30] towards two 80 mm diameter active area position sensitive channel plate detectors equipped with delay-line position readout [31]. The electron arm of the spectrometer had a 5.5 cm acceleration region. The ion arm consists of 2.2 cm acceleration and 4.4 cm drift region. The different field regions were separated by 80% transmission woven stainless steel meshes with 200  $\mu\text{m}$  mesh width. The same mesh was used about 5 mm in front of the channel plate detector on the ion side to shield the postacceleration field for the channel plate from the spectrometer drift region. The setup yields  $4\pi$  collection solid angle for electrons up to 30 eV and protons up to 16 eV kinetic energy. The resolution in  $k_p^+$  is limited by the extension of the reaction volume which is about 0.2 times the detector resolution (0.3 mm) and the internal temperature of the gas jet (about 0.5 a.u. in jet direction). 1.5 mm extension is given by the width of our gas jet. With a mean time of flight of 770 ns the protons yield 1.1 a.u./mm resolution on the detector. From these numbers one obtains an angular resolution for the sum momentum of about 10–15 deg in the polarization plane and about a factor of 2 worse for the angle to the photon beam axis. All figures presented in this paper show only the angle in the polarization plane.

The reason to choose circular polarized light for this study is to avoid any structure in the angular distribution which is due to the light polarization. Linear polarized light leads for an initial state of  $s$  symmetry to a dipole pattern, i.e. an angular distribution given by  $\cos^2$  with respect to the polar-

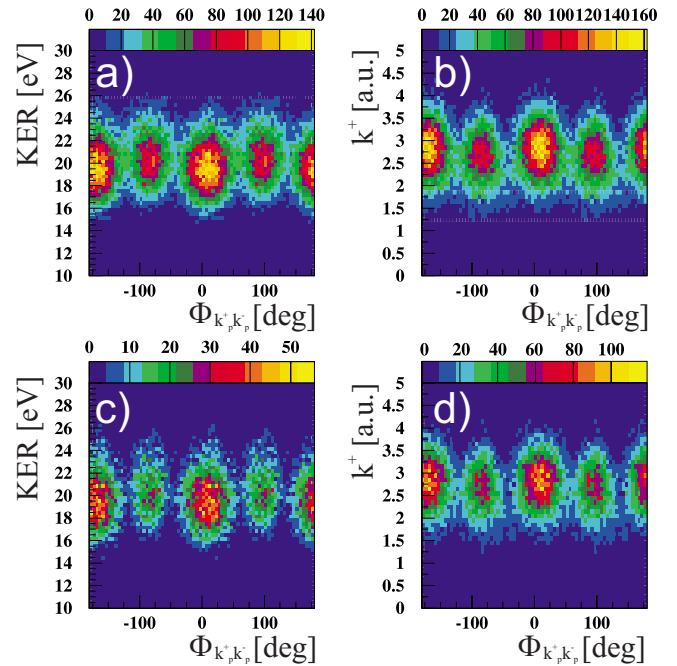


FIG. 1. (Color online) Double photoionization by 160 eV circular polarized photons. Horizontal axis: angle between molecular axis and the sum momentum of the electron pair  $k_e^+$ . Molecular axis and  $k_e^+$  are confined within  $\pm 20$  deg to the polarization plane. In (a) and (c) the vertical axis is the kinetic energy release (KER), in (b) and (d) it is the modulus of the electron sum momentum. (a) and (b) show the pattern for the whole KER range and for all sum momenta, (c) shows the same as (a) but for a restriction on the sum momenta:  $3 \text{ a.u.} < k^+ < 4 \text{ a.u.}$ , (d) shows the same as (b) with a limitation of the kinetic energy release:  $18 \text{ eV} < \text{KER} < 22 \text{ eV}$ .

ization axis. This dipole pattern would be superimposed over the diffraction pattern from Eq. (1). For circular polarized light in contrast, any atomic photoionization would yield a spherical symmetrical angular distribution in the polarization plane. Hence any structure can uniquely be attributed to a molecular effect. The dipolar structure for linear polarized light is known to prevail also as a propensity rule for double ionization of an  $s$  state. The electron momentum sum  $k_e^+$  from double ionization of helium shows a  $\beta$  parameter which is between 1 and 2 [32–35]. Double ionization by circular polarized light leads to structure in the angular distributions only if one electron is fixed (see [36] Fig. 3).

Figure 1 shows the angle between  $k_p^+ = -k_e^+$  and  $k_p^-$ , i.e. the angle between the electron sum momentum and the molecular axis versus the measured KER [Figs. 1(a) and 1(c)] and versus the modulus of the sum momentum  $k_e^+$  [Figs. 1(b) and 1(d)]. We have restricted  $k_e^+$  and  $k_p^-$  to  $\pm 20$  deg to the polarization plane. The angle shown throughout this paper is the azimuthal angle around the light propagation. The upper two panels show the full dataset, while the lower panels show a subset of the data for which KER or  $k^+$  are restricted. Clear interference minima and maxima are visible in all panels. The interference contrast is maximum in panel (c), i.e. for the largest  $k_e^+$  ( $3 < k_e^+ < 4$  a.u.). Close inspection of the figures shows that the individual peaks do not line up exactly but are alternately shifted slightly up and down in KER or  $k_e^+$ . This

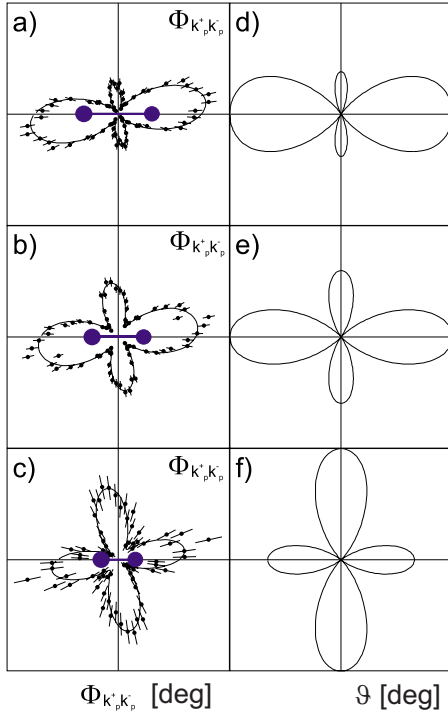


FIG. 2. (Color online) In (a), (b), and (c) different slices of Fig. 1(c) are shown to examine the KER and hence the  $R$  dependence of the interference pattern. The solid line is a guide to the eye.  $k_e^+$  as well as  $k_e^-$  are restricted in the polarization plane of the circular polarized light within  $\pm 20$  deg, the molecular axis is fixed horizontally  $\pm 10$  deg. In (a) the angular distribution is presented for  $16 \text{ eV} < \text{KER} < 18 \text{ eV}$  ( $R \approx 0.85 \text{ \AA}$ ), in (b) it is  $20 \text{ eV} < \text{KER} < 22 \text{ eV}$  ( $R \approx 0.65 \text{ \AA}$ ) and in (c) the range is  $24 \text{ eV} < \text{KER} < 27 \text{ eV}$  ( $R \approx 0.55 \text{ \AA}$ ). (d), (e), and (f) show the double slit formula of Eq. (2) with  $\beta=0.6$ .

slight shift is a result of the  $R$  and  $k$  dependence of the interference. This is much easier to see by looking at selected slices from Figs. 1(b) and 1(d) which are shown in Figs. 2 and 3.

We first examine in Fig. 2 the  $R$  dependence of the interference for a fixed value of  $k_e^+$ . The three panels show the angular distribution of  $k^+$  for three regions of the KER, which correspond to internuclear distances  $R=0.85$ ,  $0.65$ , and  $0.55 \text{ \AA}$ . The molecular axis is horizontal, the light is circularly polarized and propagates into the plane of the figure. One notes that the pattern is slightly tilted. This tilt is a result of the circular polarization of the light. We have verified that switching of the polarity inverts the tilt. This circular dichroism is well known for molecular photoionization [37–40]. The magnitude is also for H<sub>2</sub> well reproduced by theory [21]. Clearly a very strong dependence of the emission pattern on the KER is observed. For a double slit one expects that the interference fringes move closer with decreasing distance between the slits. In the present case the wavelength is so long, that we see only the first interference minimum at around  $50 \text{ deg}$  to the normal of the molecular axis. The second order maximum is already beyond  $90 \text{ deg}$  to the normal. Hence the main effect of a changed slit width at these long wavelengths is a change of the relative size of the zero and first order maximum. To verify that the observed

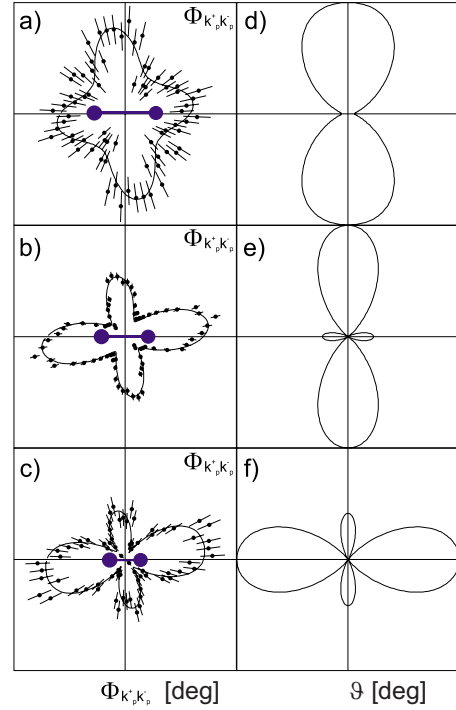


FIG. 3. (Color online) Interference pattern for different slices from Fig. 1(d) to determine its  $k_e^+$  dependence.  $k_e^+$  and  $k_e^-$  are restricted as in Fig. 2. (a) shows the slice  $1.6 \text{ a.u.} < k_e^+ < 1.8 \text{ a.u.}$ , in (b) the range of  $k_e^+$  is:  $2.4 \text{ a.u.} < k_e^+ < 2.8 \text{ a.u.}$  and in (c) the limitation of  $k_e^+$  is:  $3.7 \text{ a.u.} < k_e^+ < 4 \text{ a.u.}$ . The solid line is a guide to the eye. Panels (d)–(f) show the results of Eq. (2) for  $\beta=0.8$ .

effect is in qualitative agreement with the expectation from the simple double slit we plot the function

$$\cos \left[ k \frac{R}{2} \cos(\vartheta)^2 \right] \left[ 1 + \beta \frac{3}{2} \cos(\vartheta)^2 - \frac{1}{2} \right]. \quad (2)$$

For  $\beta=0$  this yields the double slit formula of equation (1). We have chosen  $\beta=0.6$  to yield the highest similarity with the experiments [see Figs. 2(d)–2(f)]. This modification of the optical double slit formula with the second term simulates the fact that the dielectrons are preferentially emitted along the molecular axis.  $\beta$  is the same in all three figures. This simple modeling can of course not account for the rotation of the pattern by the circular light. The only purpose of this modeling is to confirm that the observed  $R$  dependence is qualitatively in agreement with the expectation from a double slit. In turn the observed changes can also be taken as a proof that the KER measurement is indeed a direct measure of  $R$ . The observed diffraction pattern serves as an interferometric measurement of molecular structure while the Coulomb explosion imaging is a completely independent measure of the same structure.

We now test how far the double slit analogy can be taken by fixing the KER (i.e.  $R$ ) and investigating the  $k_e^+$  dependence of the interference pattern in Fig. 3. Formally in Eq. (1) varying  $R$  and  $k$  both should yield the same result. Figure 3 shows that qualitatively the pattern change in the expected direction. Again Figs. 3(d)–3(f) show the equation (2) for  $\beta=0.8$  and the parameters chosen in Figs. 3(a)–3(c). Quan-

titatively however, there is no agreement. The observed  $k$  dependence is much weaker than the one predicted by Eq. (2). We attribute this to the use of a plane wave and the complete neglect of the molecular potential. The apparent wavelength for photoelectron diffraction is known from EXAF to be considerably different from the real wavelength [41]. A good discussion of this effect for  $H_2$  can be found in [42]. On top of these known effects is the much more intriguing influence of electron correlation. In Fig. 2 we have fixed  $k_e^+$ . By this we have selected the same relative angle and energy sharing between the electrons in all panels of Fig. 2. In Fig. 3, however, we have varied  $k_e^+$  which means that different dynamical configurations are selected in each panel of Fig. 3. To account for this requires a full calculation. Such calculations are possible today [43–48]. As of today, however, none of these calculations has been extended to the high photon energies necessary to investigate interference effects.

In conclusion, we have observed interference pattern for double ionization of  $H_2$ . By measuring the kinetic energy release of the two protons we could deduce the internuclear

distance of the molecule at the instant when the electrons were removed. Our resolution in the internuclear distance  $R$  is much better than the width of the internuclear wave function  $\Delta R$  of the  $H_2$  molecule. We could therefore investigate the dependence of the interference in the electron emission on the internuclear separation. The results are in qualitative agreement with the expectation for an optical double slit.

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