

Single photon double ionization of H_2 by circularly polarized photons at a photon energy of 160 eV

Interference, decoherence and its dependence on the internuclear distance

K. Kreidi^{1,2}, D. Akoury¹, T. Jahnke¹, Th. Weber³, A. Staudte¹, M. Schöffler³, N. Neumann¹, J. Titze¹, L.Ph.H. Schmidt¹, A. Czasch¹, O. Jagutzki¹, R.A. Costa Fraga¹, R.E. Grisenti¹, R. Díez Muiño⁴, N.A. Cherepkov⁵, S.K. Semenov⁵, P. Ranitovic⁶, C.L. Cocke⁶, T. Osipov³, H. Adaniya³, J.C. Thompson⁷, M.H. Prior³, A. Belkacem³, A. Landers⁷, H. Schmidt-Böcking¹, and R. Dörner^{1,a}

¹ Institut für Kernphysik, University Frankfurt Max von Laue Str. 1, 60438 Frankfurt, Germany

² DESY, Notkestrasse 85, 22607 Hamburg, Germany

³ Lawrence Berkeley National Lab., Berkeley, CA 94720, USA

⁴ Centro de Física de Materiales and Donostia International Physics Center, 20018 San Sebastián, Spain

⁵ State University of Aerospace Instrumentation, 190000 St. Petersburg, Russia

⁶ Dept. of Physics, Kansas State Univ, Cardwell Hall Manhattan, KS 66506, USA

⁷ Department of Physics, Auburn University Auburn, AL 36849, USA

Abstract. We report on fully differential cross sections for double ionization of H_2 by a single circularly polarized photon of 160 eV energy. For an unequal sharing of the energy between the two electrons and a particular geometry where the influence of electron-electron interaction is constant, we find a four-lobe structure in the molecular frame angular distribution of the faster electron. This structure is interpreted to be due to a coherent emission of the electron from the two atomic centers of the molecule. This Young-type interference pattern is lost for other geometries, where electron-electron interaction plays a major role. Furthermore, we show that the interference structure depends strongly on the internuclear distance.

1 Introduction

Emission of electrons from a homonuclear diatomic molecule should give rise to Thomas Young-type interference effects basically due to the two center nature of the problem: on photoionization an electron wave emerges coherently from each of the two atoms of the molecule due to tunnelling of the electrons between two indistinguishable atomic sites giving rise to interference. For photo electron emission this was noted by Cohen and Fano [1] and by Kaplan and Markin [2]. The double slit analogy suggests that the interference should be visible in the angular distribution of the electron with respect to the internuclear axis of the molecule, i.e. the molecular frame electron angular distribution, giving rise to distinct differences between the gerade and ungerade parity states [3]. It has been shown however, that traces of this interference effect can even be seen in more integrated quantities like the angular distribution of the molecular breakup [1,4–6] and in the energy dependence of electrons emitted from randomly oriented molecules for ion [7–10] or photon impact [11]. Related interference patterns are also found in Auger electron angular distributions [12,13].

^a e-mail: doerner@atom.uni-frankfurt.de

The most simple and clear cut case to study this interference effect is single photoionization of H_2 (see e.g. the theoretical studies in [2,14–16]). In the present work we focus on double ionization. In this case the electronic continuum is additionally shaped by the influence of electron-electron interaction. The interplay of the interaction between the electrons with the two center interference effect is the topic of the present paper.

Experimentally the angular distribution of photoelectrons in the molecular frame is often measured by detecting the electron in coincidence with the direction of fragmentation of the molecule. This allows to use randomly oriented gas phase molecules in a measurement. This coincidence technique, however, relies on the fact that dissociation is much faster than rotation and hence the fragmentation direction coincides with the orientation of the molecular axis at the instant of electron emission (see e.g. [17,18]) which is the case for the present work on H_2 .

The present paper belongs to a series of papers in which we have studied photo double ionization of H_2 .



We briefly summarize the main findings of these preceding publications. In [19] we have reported a strong dependence of the coincident electron angular distributions on the internuclear separation, which was measured by detecting the kinetic energy release (KER) of the fragments. The photon energy was 75,5 eV resulting in only 25 eV of energy to be shared between the two electrons. The corresponding wavelength is therefore much too long to show even the first minimum of the predicted two center interference. The findings were explained [20,21] as due to a strong variation of the strength of the Σ to Π transition. In the present paper we will show an even stronger dependence of a fully differential cross section on internuclear distance. In a more recent experiment at a photon energy of 160 eV and 240 eV using circularly polarized light we found a clear two center interference [22] pattern in the angular distribution of the fast electron integrated over the emission direction of the slow electron for extremely unequal energy sharing. This interference disappears for more equal energy sharings, which we attribute to decoherence [23] induced by the now position resolving electron-electron interaction. If, however, instead of the single electron momentum k_1 the angular distribution of the sum momentum $k_+ = k_1 + k_2$ is plotted a very robust interference is found [24]. This can be traced back to a emission of both electrons from the same center of molecule [24], i.e. the ionic part of the ground state wave function. Subsequently we have shown that this interference pattern in k_+ depends on the internuclear distance [25]. In the present paper we study fully differential cross sections and report molecular frame angular distributions of one electron for a fixed angle Θ of the second electron. We show that for a geometry where the influence of electron-electron interaction is constant (i.e. the second electron is emitted perpendicularly to the plane defined by the molecular axis and the momentum vector of the first electron) the interference reappears even for an energy sharing where it is already lost for other geometries. We also show a strong dependence of the interference fringes on the internuclear distance.

2 Experiment

The experiment has been performed at the Advanced Light Source at Beamline 11 using the COLTRIMS multi particle imaging technique [26–28]. A beam of circularly polarized light ($h\nu = 160$ eV) was intersected with a supersonic molecular H_2 beam. Ions and electrons created in the intersection region are guided by a 36 V/cm electric field and a superimposed homogenous magnetic field of 14.7 Gauss [29] to two multichannel plate detectors with delayline readout [30]. Out of the measured position and the time of flight we extract the three-dimensional initial momentum vector of each particle in an offline analysis. The electric and magnetic fields and spectrometer geometry have been chosen such, that we achieve a 4π collection solid angle for electrons up to half of the maximum possible excess energy. In most cases we do not detect the higher energetic electron of the two electrons. The momentum vector of this fast electron has been deduced using momentum conservation. Since we measure the momenta of both ions and one electron the missing fourth particle momentum can be calculated. For atomic ionization this is successfully used in many experiments, for molecular targets which dissociate it is much

more challenging to reach the necessary resolution since the momentum imparted on the ions' center of mass motion by the electron pair is in the range of only 2–3 a.u. while the individual proton momentum is in the order of 40 a.u. out of the coulomb explosion. The resolution of our ion spectrometer is mainly determined by the extension of the interaction volume. Due to the very good focussing conditions of BL 11, the extension was below 0.1 mm in both directions perpendicular to the photon beam. In the photon propagation direction the length of the reaction volume was about 1.5 mm, given by the diameter of our supersonic beam. Due to the chosen geometry (see below), all angular distribution shown here are restricted to the two to the two spatial dimensions perpendicular to the photon beam.

3 Results

To search for a two-center interference pattern, we focus on a geometry where the fast electron and the molecular axis are within the polarization plane of the circularly polarized light (i.e. perpendicular to the photon propagation). In the following figures we plot the azimuthal angle ϕ_{e-mol} between the fast electron and the molecular axis, i.e. the angular distribution in the molecular frame. For the second electron we fix the corresponding polar angle Θ (i.e. the angle to the polarization plane, see Figure 1a). The virtue of this geometry in combination with circularly polarized light is, that any unaligned target species like atoms or randomly oriented molecules would yield an isotropic angular distribution in the laboratory frame. In this geometry even the two major contributions to structure in the fully differential cross section for double ionization, namely the polarization of the light and electron-electron interaction, both by themselves do not produce any structure. For all data points in Figures 1 and 2 the electron-electron interaction is identical and hence does not cause any anisotropy. Also the use of linearly instead of circularly polarized light would produce additional and for the present purpose unwanted structure by selection rules [31,32].

In Figure 1e, we observe a pronounced four lobe pattern for the fast electron, which is slightly rotated clockwise. We suggest that this pattern is the result of the double slit like interference. For two coherent spherical electron waves with momentum k_e emitted from two centers being separated in space by R one obtains a molecular frame angular distribution given by

$$I(\Phi_{e-mol}) \propto \cos(k_e R / 2 \cos(\Phi_{e-mol})) \quad (2)$$

where Φ_{e-mol} is the angle between electron momentum vector k_e and the molecular axis. To compare this equation for the optical case with the situation of an electron emerging from a molecule, additional effects like the scattering of the electron at the neighboring center and phase shifts in the molecular potential [33] have to be taken into account. These additional effects significantly change the apparent internuclear distance (a well known effect in EXAFS [34]) or alternatively the apparent electron momentum. Multiple scattering [35] as well as RPA calculations [36] for single electron emission which both include such effects show a four lobe interference structure at the current electron wavelength (Figure 1f). Please note that the calculations treat the emission of only a single active electron, while in our experiment both electrons are ejected. The tilt of the pattern is a result of the helicity of the circularly polarized light. We have verified that the rotation flips for inverted polarization. This circular dichroism in molecular double ionization has in general two origins. Firstly it is produced by the multiple scattering of the electron wave at the two centers, a well studied effect in single electron emission [37–40]. This effect is accounted for in the single electron calculations shown in Figure 1f. Secondly in double ionization also the two electrons can give rise to circular dichroism even for the atomic case [41–44]. This latter effect is excluded in the present data by the choice of the geometry, where the slow electron is emitted along the photon direction [45].

Figure 1e shows, that for the chosen geometry the presence of the second electron does not destroy the interference. If, however, no constraint on the angle of emission of the second electron is made (Figure 1b) the interference is completely lost. As discussed in [22] this loss of fringe contrast is analogous to the decoherence in matter wave interference experiments, where a scattering [46,47] or emission [48] of photons or a collision with other atoms [49] behind a

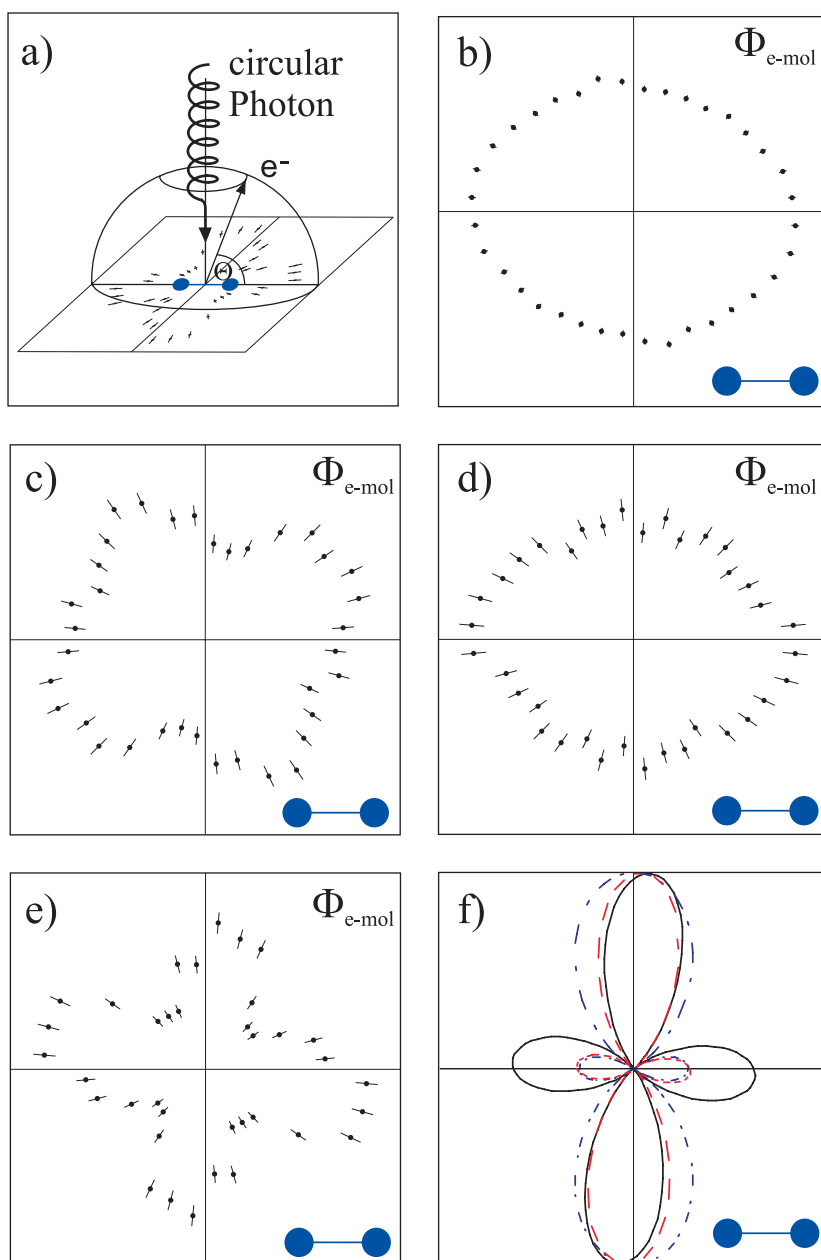


Fig. 1. Double photoionization by 160 eV circularly polarized photons. a) Demonstration of the geometry chosen in this paper. b) Polar presentation of the azimuthal angle Φ_{e-mol} between the fast electron ($85 \text{ eV} < E_1 < 105 \text{ eV}$) and the molecular axis in the polarization plane. The polar angle of the molecular axis and the fast electron is fixed in this plane $\pm 20^\circ$. The molecule is aligned horizontally. There is no angle constraint for the slow electron ($5 \text{ eV} < E_2 < 25 \text{ eV}$). c)–e) Same as b) but with a constraint for the polar angle θ of the slow electron: c) $\theta = 0^\circ\text{--}6^\circ$, d) $\theta = 37^\circ\text{--}45^\circ$ and e) $\theta = 75^\circ\text{--}90^\circ$ with respect to the polarization plane. In b)–e) there is no constraint on the azimuthal angle of the slow electron. f) Angular distribution of a single electron with the kinetic energy E_1 . Red (dashed) curve: equation (2), blue (dashed dotted) curve: RPA calculation, black (solid) curve: multiple scattering calculation.

double slit arrangement, resolving this two-centre setup, yields a similar loss of fringe contrast. This can be understood as the result of a phase shift or a momentum transfer with a de Broglie wavelengths shorter than the bond lengths in the in the collision between the particles. Such a collision leads to an entanglement of the two particle wave function. In the case of single photon double ionization electron-electron collisions within the target are the dominant ionization mechanism within high photon energies and the energy sharings employed in this experiment ([50,51]). One can also argue, that the electron-electron interaction deposits which way information in the second particle, again leading to a loss of contrast. Figure 1 shows, that a restriction of the opening angle into which the second electron is ejected recovers the interference. This finding can be discussed in different contexts. Firstly thinking about phase shifts on the wave function of the fast electron induced by the slow electron, the selection of the direction into which the slow electron is scattered (Figure 1) determines this phase shift. For the particular out of plane geometry as in Fig. 1e, the phase shift induced on the two partial waves from the two centers is equal and hence this figure is closest to the unperturbed single electron diffraction as in the calculations. In contrast, the selection made in Figure 1b includes all in plane directions of the slow electron. This results in an averaging over many different phase shifts, which obscures the interference completely. Another way to rationalize this finding is to think of the momentum measurement on the slow electron into a fixed direction as a quantum eraser. The two electron emission creates an entangled electron pair in which in principle one electron can be used to infer which way information on the second electron. By measuring the momentum of this electron one erases all such position information. To continue this argument the alternative choice to erase position information by momentum detection would be to actually use the slow electron for a position measurement. In this case one would have to focus the slow electron wave with a wide opening angle into a photoelectron microscope (PEEM). Such an experiment is unfortunately technically not yet feasible today.

To further support our claim that the observed structure in Figure 1e results from the two center interference we investigate its dependence on the internuclear distance R in Figure 2. For each event our experiment measures the kinetic energy release KER with high resolution (<500 meV). In a classical picture of the nuclear motion the KER is inversely proportional to the internuclear distance at the instant of double ionization [19,52–54]. We have previously demonstrated that this technique of Coulomb explosion imaging for double ionization is precise enough to distinguish even between a nuclear wave function of a harmonic and a Morse potential (Figure 1 in [19]). Figure 2 shows the data from Figure 1e split in three subsamples to different KER. We find a striking dependence on the internuclear distance. To verify that the observed effect is in qualitative agreement with the expectation from the simple double slit we plot the function

$$\cos(kR/2 \cos(\vartheta))^2 \cdot \left(1 + \beta \left(\frac{3}{2} \cos(\vartheta)^2 - \frac{1}{2}\right)\right). \quad (3)$$

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 experiment. This modification of the optical double slit formula with the second term simulates the fact that the electrons are preferentially emitted along the molecular axis. β is the same in all three figures. We have used an apparent $k = 3.1$ a.u. This simple modelling can of course not account for the rotation of the pattern by the circular light. The only purpose of this modelling is to confirm that the observed R dependence is qualitatively in agreement with the expectation from a double slit.

We note that Horner and coworkers [20] have shown theoretically the relative strength of the σ - and π -transition in double ionization does significantly change as function of R . This finding has recently been beautifully confirmed in [21]. It is currently not clear how this finding connects to the observation shown in Figure 2 with our seemingly different interpretation based on interference.

To conclude we have demonstrated a double slit interference pattern in the fully differential cross section for double photo ionization of H_2 for a special geometry where the

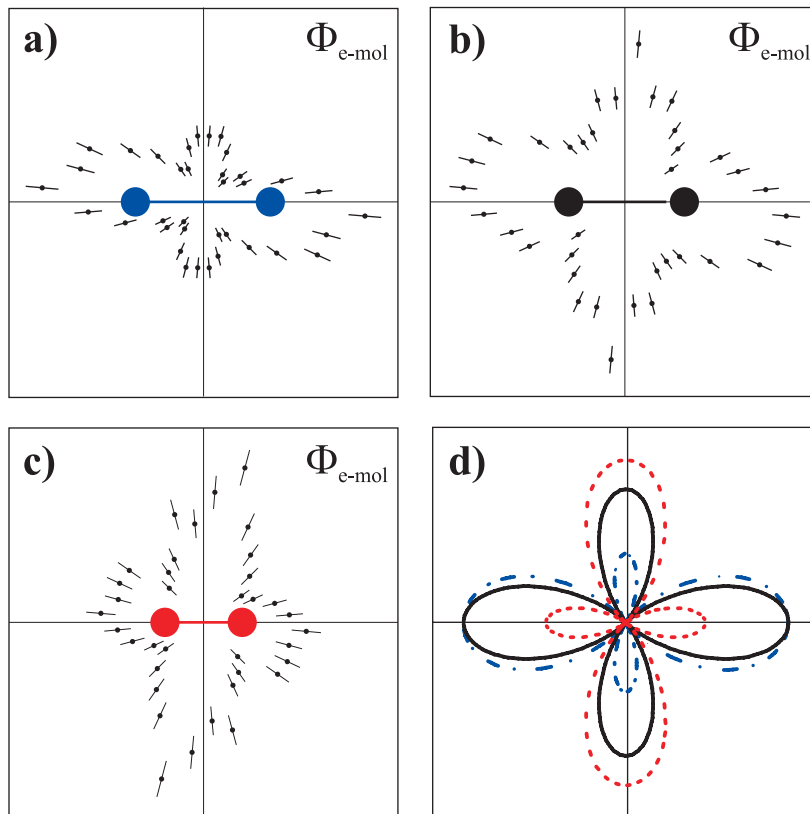


Fig. 2. a)–c) Angular distribution of the fast electron for the geometry chosen in Figure 1e but with a variation of the KER and hence the internuclear distance R , a) KER = 15 eV–18 eV ($R \approx 0.85$ Å), b) KER = 19 eV–21 eV ($R \approx 0.7$ Å), c) 22 eV–24 eV ($R \approx 0.6$ Å). d) Double slit formula according to equation 3 with $\beta = 0.6$ for different internuclear distances R , blue (dashed dotted) curve: $R = 0.85$ Å, black (solid) curve: $R = 0.7$ Å, red (dotted) curve: $R = 0.6$ Å.

influence of electron-electron interaction is constant. The interference fringes are washed out for other geometries and are shown to depend strongly on the internuclear distance. While original pioneering theoretical work on photo double ionization of H_2 , for example in the 5C approach, has been reported e.g. in [14], today quasi exact calculations for photo double ionization of H_2 within the fixed nucleus approximation have been reported by two groups [20,21,55–59]. Since increasing photon energy requires a larger angular momentum basis these calculations have so far not been extended to higher photon energies where interference phenomena become important. Our data show that such calculations would be highly rewarding. Examination of the two body wave function obtained from such calculations promises to build a bridge between many particle quantum dynamics and quantum optics.

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