Transition from random to oriented target properties in molecular hydrogen

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Abstract

The analogy of homonuclear diatomic molecules to the Young type double slit experiment such as Cohen-Fano cross section oscillations [1] has been topic of a large amount of photoionization investigations. The two center interference is caused by electron non-locality in homonuclear molecules. Here, the hydrogen molecule's relative cross section oscillations are analyzed over a large (29eV-1200eV) photon energy range distinguishing between the randomly in space distributed molecule and the 'oriented sample' in which the orientation of the molecular axis to the polarization light vector is determined. Furthermore, a transition effect from random to oriented target properties is detected when the de Broglie wavelength of the photoelectron resolves the internuclear distance. The ion fragment angular distribution given by the $\beta_{_{\rm m}}$ parameter is analyzed, compared to data of Kossmann et al. [2] with good agreement and found as a reason for the transition. The β_m value converges to -1 at high photon energies resulting in a preferred molecular orientation for the ionization process. The similar transition behavior is observed in the inner shell ionization data of also tunable by the photoelectron's de Broglie wavelength. Therefore, several variants of ion time of flight detectors (position sensitive, pure time of flight) spectroscopy, different drift tube lengths) with the option of coincidence mode with an electron time of flight spectrometer were used. The experiments were performed at BESSY II (U125/2, UE56/1-PGMb, UE56/2 PGM1) and PETRA III (P04) facilities.

Spectra analysis



Figure 1: Schematic representation of the molecular cross section oscillation: the caused by the interference of indistinguishable H-atoms oscillation, which is analogous to the double slit experiment, takes place along the atomic cross section.



Figure 2: β_m as a function of photon energy combined with a fit of a transition function: For high photon energies the angular distribution converts to -1 which results in an anisotropic distribution. Low energy data of Kossmann et al. [2] can be continued with our data.



Figure 6: Ion angular distribution analysis: The time of flight spectra can be used to analyze the orientation of the molecule to the polarization vector for (H^+, H^+) -ionization. Due to Coulomb explosion the orientation has a strong influence to the ion time of flight.

Figure 3: Average ion fragment angular distributions before (blue) and after (red) the transition presented in figure 2 and 5. For better comparison, the high energy data (red) is mirrored on y-axis. An isotropic distribution is also shown in black



Figure 4: ASame transition behavior is observed in N₂ inner shell ionization in the data of Zimmermann et al. [5]. The relative cross section oscillations of σ_{g} and σ_{u} with a transition (black) similiar to H₂.





Figure 5: U-shape angular distribution analysis of the H⁺-feature: The spectra are normalized to the total intensity of the full H⁺-intensity for each photon energy. In addition, the β_m parameter and a time of flight spectrum are shown. The transition can be observed directly.





Figure 7: Ion time of flight spectra of the H⁺ feature (blue dots) at 185eV in a combined view with the u-shape-fit (red curve) and central peak fit (black curve). The time of flight channel resolution is 120ps.

PETRA III P04

Recently we were able to expand our data range at PETRA III P04 beamline up to 1200eV, analysing σ oscillations.



Figure 8: Our experiments were performed at BESSY II facilities at beamlines U125/2 for lower photon energy, UE56/1-PGM for energies up to 530eV and at PETRA III P04 up to 1200eV.

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Figure 9: 1000eV ion spectrum (measured with the setup of S. Deinert)

Figure 10: Relative cross section oscillations up to 1200eV: Hydrogen molecule's Cohen-Fano oscillations of the random (low energy data from [3] in magenta normalized to atomic cross section [4]) and oriented target. The expected 90° phase shift is observed at low photon energies.

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