Attosecond coincidence spectroscopy of diatomic molecules

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Abstract. : Sub-cycle ionization of Ar_2 by few-cycle laser fields is investigated with COLTRIMS. Low energy photoelectrons show clear deviations from double slit interference. We suggest that breakdown of the single-active electron approximation could be responsible for such effect.

Introduction

Recent attosecond experiments have been based on precise GV/cm electric field synthesis (see e.g.[1]). Few cycle fields have been produced via control over the carrier-envelope offset phase in combination with enhanced spectral broadening and mirror compression techniques. With such techniques single events of strong field tunneling have been timed with sub-cycle accuracy [2]. Sub-femtosecond ionization has become an attractive tool for time-resolved spectroscopy, in particular via rescattering of the liberated sub-femtosecond electron wavepacket onto the parent molecule, and via attosecond XUV pulse generation from HHG, which is a well-known daughter process of rescattering [3]. Attosecond XUV pulses have been used for attosecond time domain spectroscopy, but presently only for a limited number of text-book cases (see e.g. [4, 5]). Successful application of attosecond spectroscopy to molecular systems is pending for many cases. In our recent experiment we have applied sub-cycle ionization to systems consisting of two identical atoms (Argon dimers), thereby asking for geometrical information about the system via the interference pattern of the outgoing electrons. In principle this information could be retrieved with attosecond timing accuracy. We have expected that the photoelectron energy spectra would be fully determined by two-center interference. We found that this is only partially true.

Experimental Methods

To investigate the interference effect in strong-field ionization of molecules we have used a reaction microscope. We have applied it here to measure photoelectron momentum distributions in coincidence with Ar^+ and Ar_2^+ ions. Ar_2 is usually present to an amount of a few percent in supersonic expansions of Argon. The internuclear separation of the Van-der-Waals bound dimers is distributed from 6.8 to 9.4 Å. After collimation the supersonic beam crosses a Ti:sapphire laser beam in its focal spot where an intensity of 2×10^{14}W/cm^2 is reached. The laser pulses have a FWHM of 5.5 fs and a central wavelength of 770 nm. A weak electric field (7.5 V/cm) is applied across the interaction region to extract photoelectrons and ions. After acceleration and subsequent

field-free drift ions and electrons are detected by two position sensitive multichannel plate detectors. A homogeneous magnetic field of 8.5 Gauss parallel to the electric field additionally guides photoelectrons to the detector. For each individual particle the time-of-flight is recorded along with the position where it hits the detector. From such data the full momentum vector for each particle can be reconstructed.

Results and Discussion

Fig.1 shows a comparison between the photoelectron momentum distributions (in cylindrical coordinates) for Ar and Ar_2, with our theoretical simulations. In our model we derive the molecular momentum distribution $|M_{A-A}(\mathbf{p},\mathbf{R})|^2$ based on the atomic momentum distribution $|M_A(\mathbf{p})|^2$ multiplied with appropriate Franck-Condon factors and additional R-dependent double slit interference terms. The latter depend on the respective atomic orbitals from which electrons can be removed ($3p\sigma_u$, $3p\pi_u$, $3p\pi_g$). The calculation is based on strong field approximation (SFA) to the transition state matrix to the continuum. In particular, three final states can be accessed and lead to stable Ar_2^+. Least square agreement with the experimental data can be reached when using a weighted superposition of these three transitions at an internuclear distance of about 7.4 Å. For non-aligned molecules we calculate momentum distributions according to

$$\begin{aligned} |M_{A-A}(\mathbf{p},\mathbf{R})|^2 = \; & 0,11\cdot|M_A(\mathbf{p})|^2\left|S_{fi}^{3p\sigma_u}\right|^2\left[\tfrac{1}{6}+\tfrac{1}{2}\int\tfrac{d\Omega_{\hat{\mathbf{R}}}}{4\pi}\cos(\mathbf{pR})\cos^2\theta_R\right] \\ & +0,57\cdot|M_A(\mathbf{p})|^2\left|S_{fi}^{3p\pi_u}\right|^2\left[\tfrac{1}{3}+\tfrac{\sin(pR)}{2pR}-\tfrac{1}{2}\int\tfrac{d\Omega_{\hat{\mathbf{R}}}}{4\pi}\cos(\mathbf{pR})\cos^2\theta_R\right] \\ & +0,32\cdot|M_A(\mathbf{p})|^2\left|S_{fi}^{3p\pi_g}\right|^2\left[\tfrac{1}{3}-\tfrac{\sin(pR)}{2pR}+\tfrac{1}{2}\int\tfrac{d\Omega_{\hat{\mathbf{R}}}}{4\pi}\cos(\mathbf{pR})\cos^2\theta_R\right] \end{aligned} \quad (1)$$

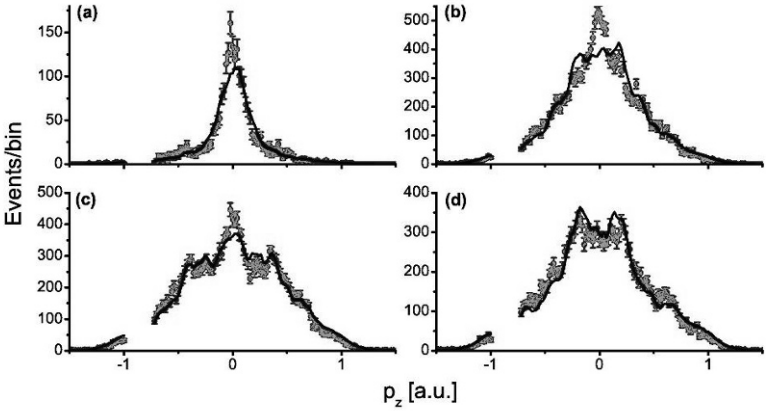

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Fig. 1. : Least square fit (full black line) of a sum of simulated Ar_2 momentum distributions to the measured Ar_2 photoelectron momentum distribution (gray dots with error bars). Cuts shown are made at the radial momenta $p_r = 0.01 \pm 0.01$a.u. (a), $p_r = 0.05 \pm 0.01$ a.u. (b), $p_r = 0.09 \pm 0.01$ a.u. (c), and $p_r = 0.15 \pm 0.01$ a.u. (d). The least square fit was made for graph (c). The simulated spectra for all other cuts use the same fit parameters as in (c).

It can be seen that the experimental data deviate from theory significantly only for small electron momenta. We interpret this as a manifestation of an energy exchange between the outgoing tunnelling electron and the remaining Ar_2^+. If timescales between departure and typical charge oscillations in the remaining system are comparable, deviations from a double slit interference could become likely. Moreover, intramolecular recollisions during ionization are not taken into account within our SFA model. Such recollisions could be 10-20 times more relevant for dimers than for isolated atoms, and could lead to internal excitation, thereby obscuring interference.

Conclusions

Our experiment suggests that directly emitted electrons can be used to retrieve information about emitter geometries on attosecond timescales. However, we find that particle momenta should be high enough to avoid energy exchange during the ionization. The observed low momentum deviations appear to be a clear manifestation of a failure of the single-active electron approximation for tunnelling, and it is quite interesting that they can be observed even in physically quite well separated systems like Van-der-Waals clusters. Because sub-cycle tunnelling is a basic ingredient for most attosecond experiments, breakdown of SFA close to threshold ATI may affect the interpretation of various future molecular attosecond experiments. Our result also adds support to our previous observations that non-adiabatic multielectron effects in molecular strong field ionization should not be neglected [6].

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