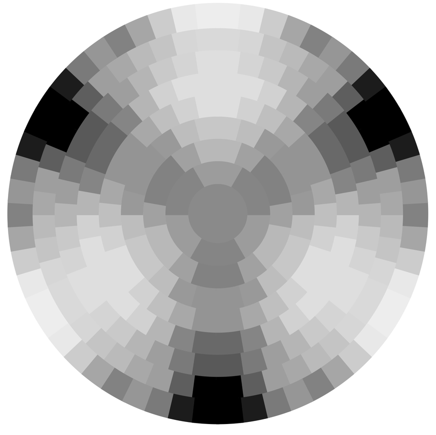

Electric field induced predissociation of high Rydberg-states in H_3 and D_3

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We examine the **decay** of the isotopologues H_3 and D_3 into three hydrogen atoms. Collecting temporal and spatial information of all fragments, allows us to deduce the full **momentum-vector-correlation** of such a decay. The probability-distribution is represented in so-called Dalitz plots and sensitively depends on the rovibronic level of the decaying molecule.
Here we present results on the 3-body-decay of **high Rydberg-states [5]** which are induced by **Stark-effect** via a localized electric field of variable strength. We find that the Dalitz plots of these states in H_3 rapidly change with electronic energy. In the immediate vicinity of the ionization threshold we observe a **preferred dissociation in near linear configuration**, where in the center-of-mass frame one atom stays at rest while the remaining two gain opposite momenta. This finding is **similar to observations by Strasser *et. al.*** [6], who studied dissociative recombination of cold H_3^+-ions with slow electrons in a complete different experiment.
Very recently we have started to perform measurements in D_3. Preliminary results are shown below.

1.Introduction to H_3

- H_3 is the **most simple** neutral polyatomic molecule (**3p + 3e**) and serves as a **prototype system** for theoretical molcecule-dynamics.
- Despite of its simplicity, H_3 shows **complex effects** such as **non-adiabatic couplings** which lead to **predissociation**.
- The **groundstate** of H_3 ist **doubly degenerate** in equilibrium configuration and **dissociative**, which means that H_3 is an **excimer**. Propagation on the groundstate-surfaces leads ultimatively to the two **decay-channels**:
  $H_3 \rightarrow H(1s) + H_2(\nu,J)$ or **$H_3 \rightarrow H(1s) + H(1s) + H(1s)$**
- With exception of a **metastable level** $2pA_2''$ (τ = 640ns) all low-lying states are subject to **strong non-adiabatic couplings** to the ground-state which limit lifetimes to a maximum of order ~10ns. **Rydberg- states** higher than n=5 can again show longer lifetimes.

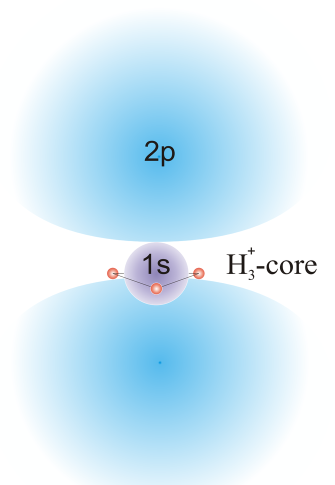

Fig.1 *Orbital of the metastable $2pA_2''$-level*

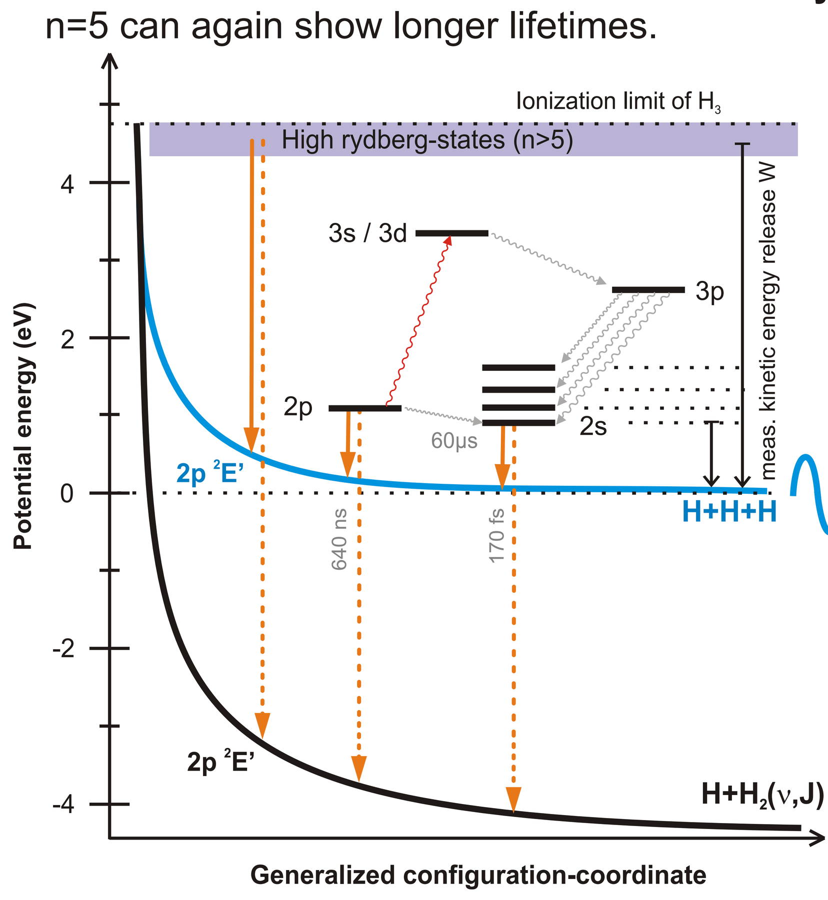

Fig.2 *Simplified level-scheme of H_3*

$$x = 3(\varepsilon_2 - \varepsilon_1)/\sqrt{3}$$
$$y = 3(\varepsilon_3 - 1/3)$$

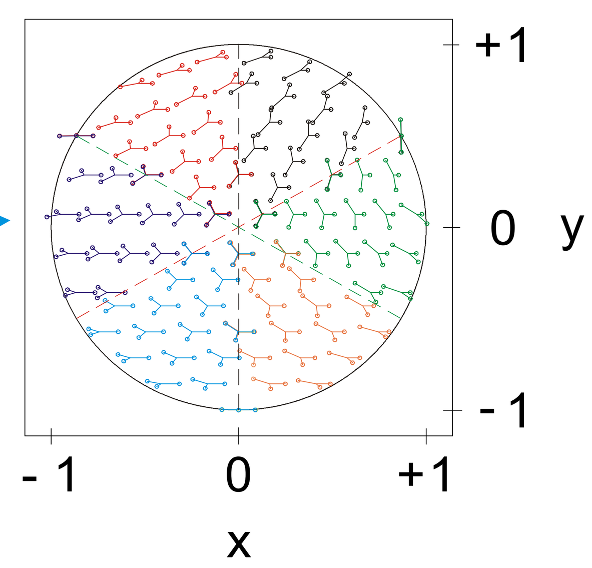

Fig.3 *Representation of momentum-vector-correlation: the Dalitz plot*

2. Experimental setup

- Formation of neutral H_3 in different levels via **charge-exchange** of 3keV H_3^+-ions with cesium-atoms ($H_3^+ + Cs \rightarrow H_3 + Cs^+$)
- **deflection** of non-neutralized ions by means of a weak electric field
- molecules in **short-lived** states dissociate before they pass an aperture 300mm downstream and remain **undetected**
- **long-lived** states can enter the dissociation-region where they can be subject to an inhomogeneous, static but very localized **electric field** (max. 36 kV/cm) or are excited to higher levels by a **laser**

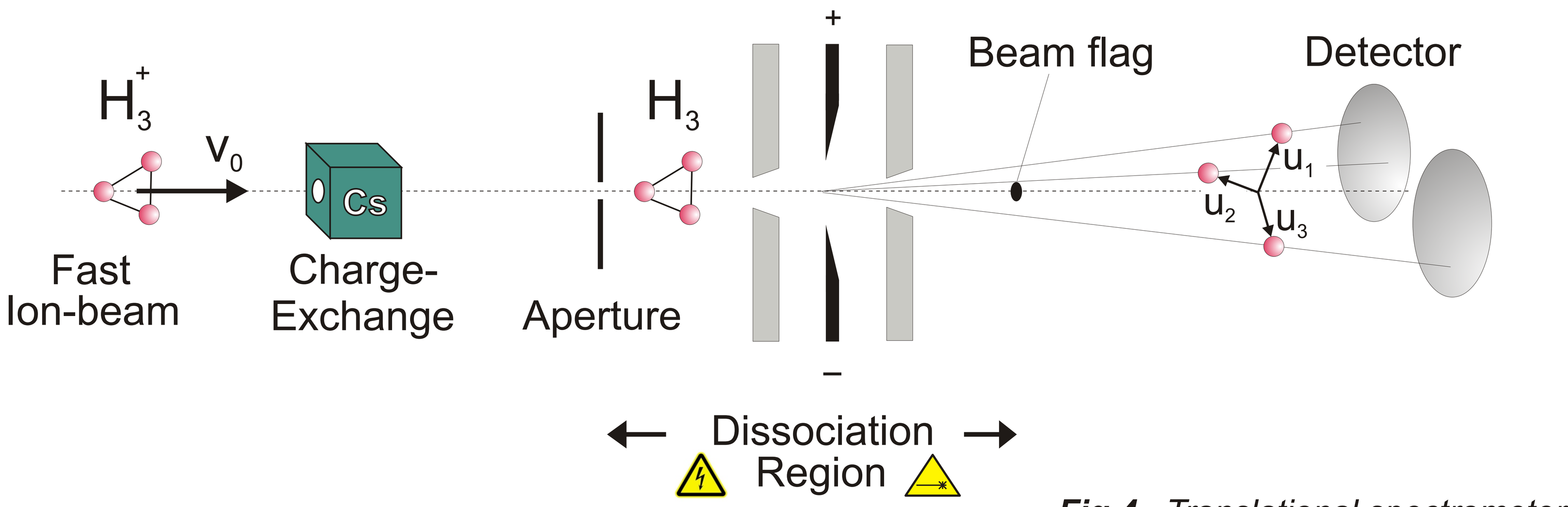

Fig.4 *Translational spectrometer*

- dissociation fragments with sufficient transverse momentum pass the beam flag and impinge on the **time- and position-sensitive detector** (MCPs and delayline-anodes)
- identification of correlated fragments via several **coincidence criteria**
- from the data we deduce the **kinematically complete information** on the 3-body-decays, e.g. the **total kinetic energy release W** and the c.m. **momentum vectors** of all fragments

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3. Electric field induced dissociation

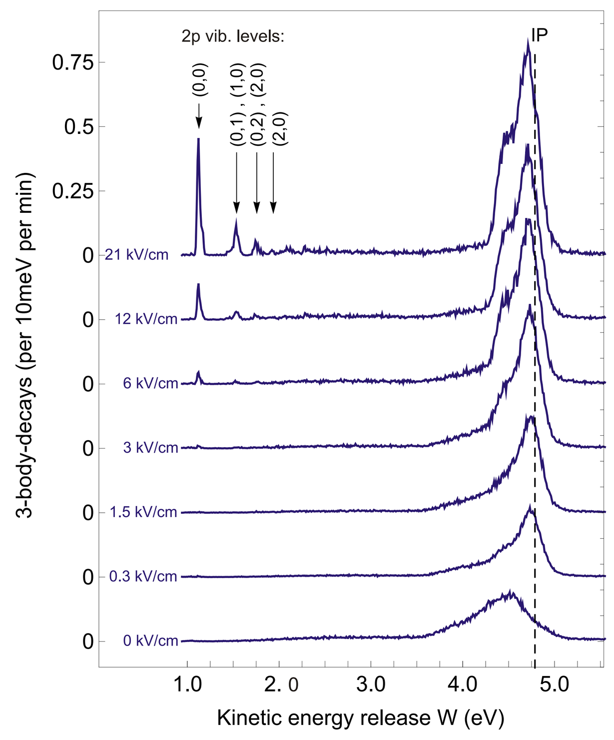

Fig.5 *Kinetic energy release spectra of H_3 at different field strengths*

- 0kV: **broad distribution** of 3-body-decays with high energies corresponding to long-lived states with n>5, which decay on their ~10cm-way between aperture and beam flag
- **increased decay-rate** with increasing field: states are mixed with rapidly predissociating levels
- the observed **distribution sharpens** as a result of the better localized space of dissociation
- the field also mixes short-lived 2s character to the 2p metastable state in different vibrational levels leading to a rapid rise in rate at **discrete energies ~ (1eV - 2eV)** [4,7]

- we have performed a **simulation** of the spectrum at 7kV in order to clarify which states contribute to the 3-body-decays observed
- just before the Stark field region, the population of molecules in a state with quantum numbers (n,l) is governed by the product of the probability $P_1(n)$ that the state is populated in charge exchange and the probability $P_2(n,l)$ of surviving the time-of-flight t from charge exchange to the field zone, we set:

$$P_1(n) = c / n^3$$
$$P_2(n,l) = \exp[-t / \tau(n,l)]$$

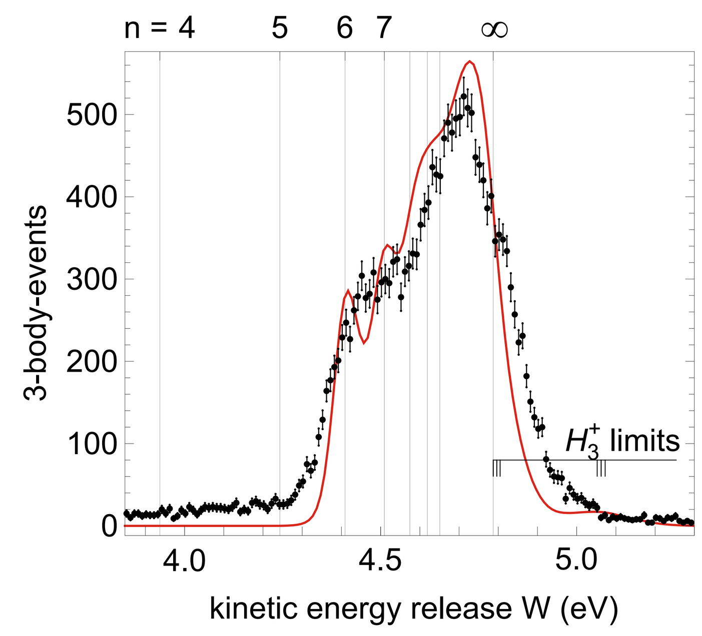

Fig.6 *Comparison of measured kinetic release spectrum at 7kV with simulation*

- we assume that only electronic states (n,l=n-1) which have the longest lifetimes contribute and take $\tau(n,l=n-1)=(n^3/350)$ µs as a scaling rule, which reproduces the **hydrogenic lifetimes**
- for the **relative vibrational population** we assume a value of 2% in the two vibrational normalmodes with respect to the vibrational groundstate
- for the **rotational population** we assume a temperature of T=1000K
- **only free parameter** is the constant c which controls the overall count rate
- each peak resulting from a rovibronic state is **folded with the experimental width**
- the simulation (red line in Fig.6) shows **good agreement with measurement**

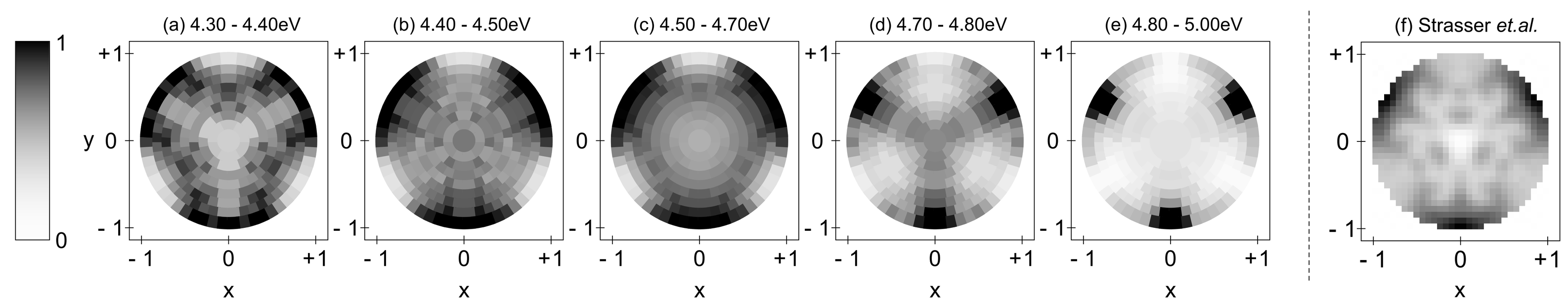

Fig.7 *Dalitz plots of decays within distinct energy-intervals near the ionization-limit of H3: (a) n=6 ,(b) n=7 , (c) 8<n<16 , (d) n>16 and 20meV above ionization-limit , (e) levels built on rovibrational excited cores,from [5]. Accumulated data for field-strengths >0 kV/cm. (f) Comparison with results by Strasser et al. at TSR [6].*

- In Fig. 7 one sees the Dalitz plots from decays in different energy-intervals within the broad distribution, which belong to distinct main-quantumnumbers. There is a **clear trend** in the development of the Dalitz in direction to **linear decay-geometry** with increasing kinetic energy release
- Dalitz plots in Fig.7 (c) and (d) bear **close resemblance** to results by Strasser *et.al.* [6], obtained in **disscociative recombination** of H_3^+ with slow electrons at the **Test Storage Ring (TSR)** in Heidelberg
- Both **entirely different experiments** observe 3-body-decays from states near the ionization-limit and reveal a **preferred dissociation in linear geometry**

5. Preliminary results on D_3

- Very recently we have started measurements of high Rydberg-states near the ionization limit of D_3
- For these measurements we changed the experimental settings to a configuration optimal for D_3
- In contrast to H_3 it seems like there are long-lived states starting with principal quantum number n=4, which have lifetimes bigger than ~700ns to survive the transition time from charge exchange to the dissociation region
- Again we are able to enhance the dissociation-rate of high Rydberg-states by admixing short lived-character via an electric field
- With current settings we are able to resolve peaks originating from decays out of states with principal quantum numbers 4,5 and 6

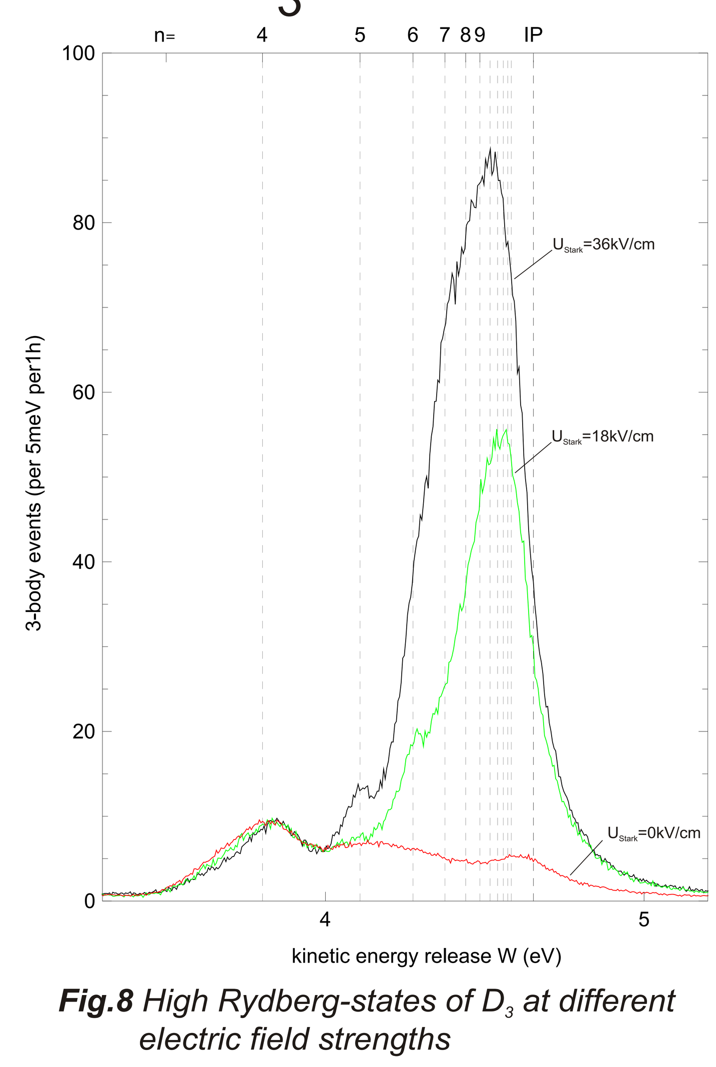

Fig.8 *High Rydberg-states of D_3 at different electric field strengths*