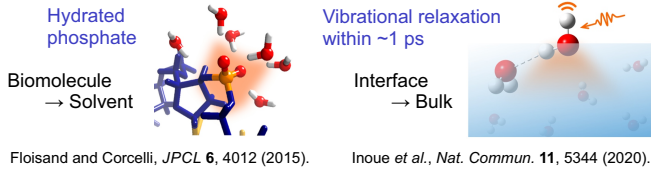


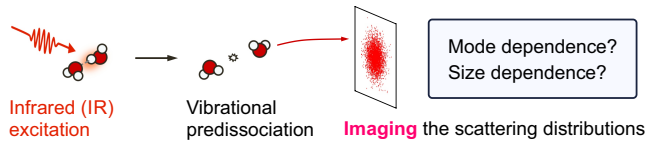
## Introduction

### Vibrational energy transfer in the condensed phase

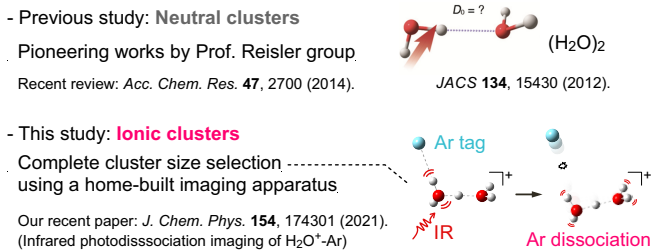


### Intermolecular vibrational dynamics

### Gas phase molecular cluster as a microscopic model system



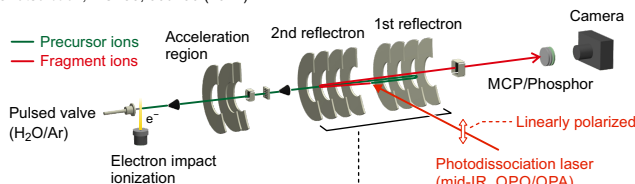
### Ion imaging experiments for infrared predissociation



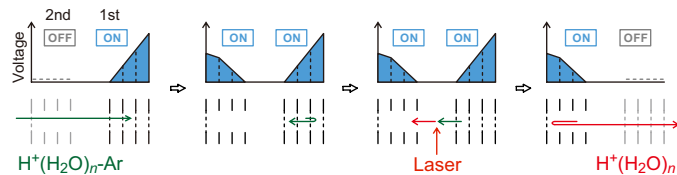
## Experiment: Photofragment ion imaging

### Imaging apparatus with a linear tandem reflectron TOF mass spectrometer

Okutsu et al., *RSI* **88**, 053105 (2017).



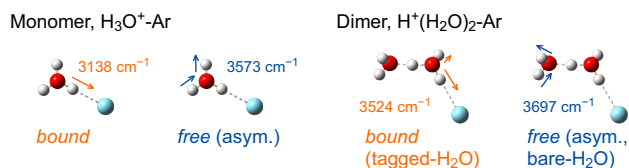
### Mass selection by pulsed high voltages



## This study

### Infrared photodissociation imaging of $\text{H}^+(\text{H}_2\text{O})_{1,2}\text{-Ar}$

IR spectra were reported in Headrick et al., *Science* **308**, 1765 (2005).

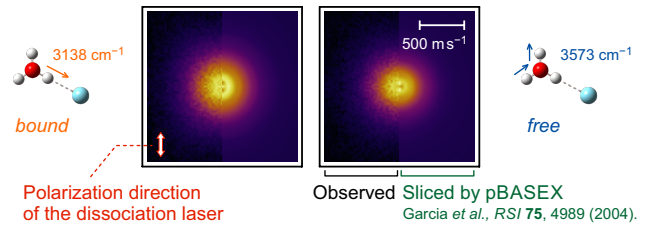


- Mode dependence? *bound*- vs. *free*-OH stretching excitation

- Size dependence?  
Any effect of an additional  $\text{H}_2\text{O}$  molecule?

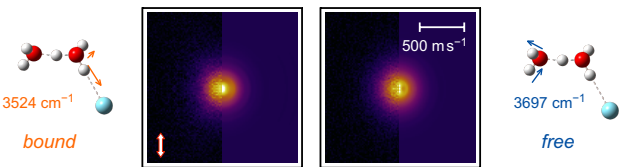
## Results and discussion

### Imaging result 1: $\text{H}_3\text{O}^+\text{-Ar}$



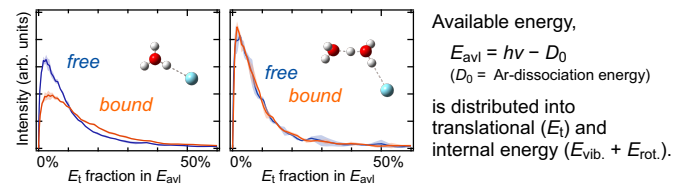
- Isotropic → Long predissociation lifetime (> Rotational period, ~10 ps)
- *Bound* excitation gave a slightly larger distribution.  
(See the following  $E_t/E_{\text{avil}}$  plot for a clearer comparison)

### Imaging result 2: $\text{H}^+(\text{H}_2\text{O})_2\text{-Ar}$



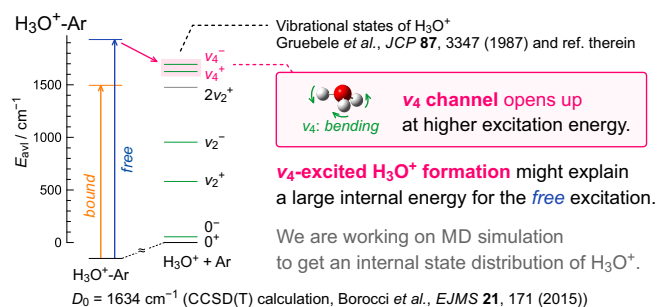
- Similar small images were observed for both modes.  
→ Translational energy did **NOT** depend on the excited vibrational modes.

### $E_t/E_{\text{avil}}$ plot: Energy fraction into the translational energy ( $E_t$ )

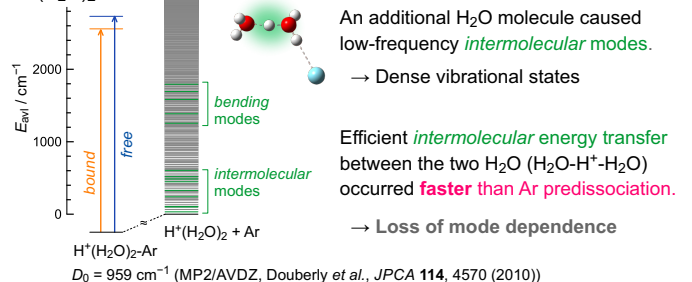


- $\text{H}_3\text{O}^+\text{-Ar}$ : *bound* > *free* → **Large internal energy** in  $\text{H}_3\text{O}^+$  for the *free* excitation
- $\text{H}^+(\text{H}_2\text{O})_2\text{-Ar}$ : *bound* = *free* → Little mode dependence in the  $E_t$  fraction

### Energy diagram for vibrational predissociation



### $\text{H}^+(\text{H}_2\text{O})_2\text{-Ar}$



## Conclusion

### Imaging study for the IR predissociation of mass-selected ions

- 1)  $\text{H}_3\text{O}^+\text{-Ar}$ : **Mode-dependent  $E_t$  distributions** due to its discrete energetics
- 2)  $\text{H}^+(\text{H}_2\text{O})_2\text{-Ar}$ : **Fast intermolecular energy relaxation** before predissociation