Supporting Information for

Highly Efficient Pumping of Vibrationally Excited HD Molecules via Stark-induced Adiabatic Raman Passage

Tao Wang[1], *Tiangang Yang*[1,2], *Chunlei Xiao*[1,*], *Dongxu Dai*[1] *and Xueming Yang*[1,*]

1.State key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, 457 Zhongshan Road Dalian 116023, China

2.School of Physics and Optoelectric Engineering,Dalian University of Technology, Dalian, Liaoning 116023,China

* Correspondence Authors: All correspondence should be addressed to the following authors: Chunlei Xiao: chunleixiao@dicp.ac.cn; Xueming Yang: xmyang@dicp.ac.cn.

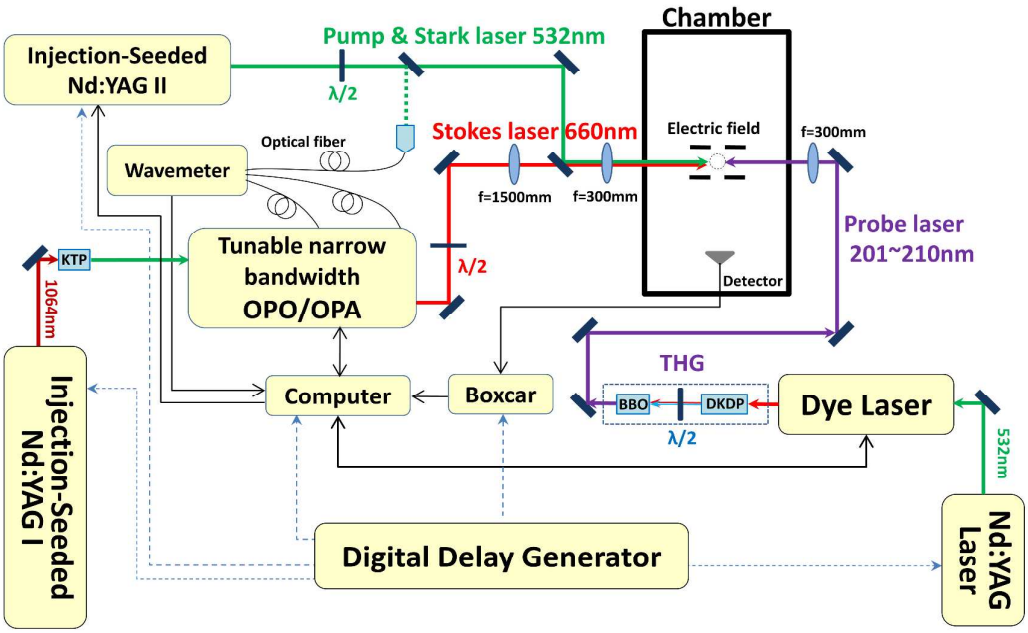

Figure S1 The schematic diagram for the laser system and the experimental setup

for the SARP experiment. OPO/OPA: optical parameter oscillator/amplifier. λ/2: half-wave plate. THG: third harmonic generation.

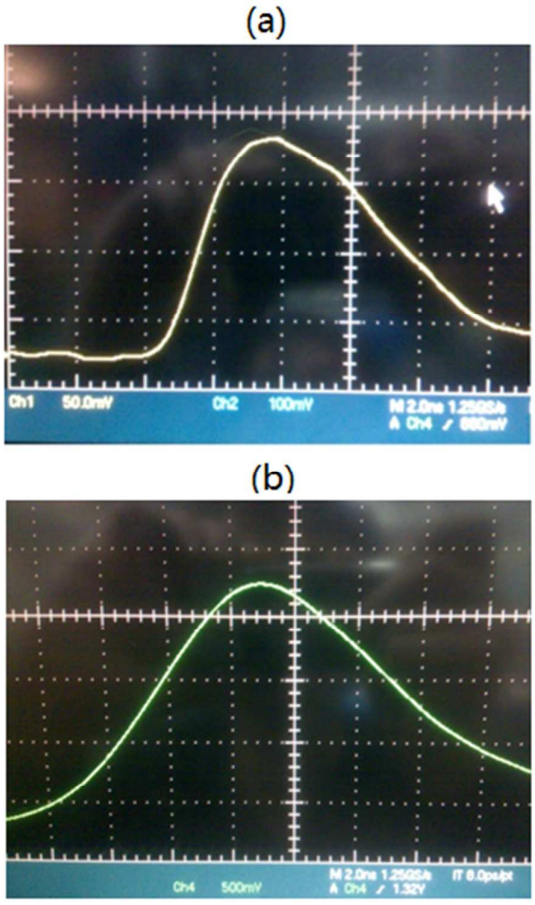

Figure S2 (a)The temporal profile of Stokes pulse, generated by a home-built optical parameter oscillator/amplifier system. (b) The temporal profile of the pump pulse, generated as the second harmonic of a seeding-injection Nd:YAG laser. They were measured with a fast photodiode and oscilloscope. Time scale is 2ns/div. It is clear to see that the OPO/Amplifier system pulse for the Stokes laser is very asymmetric, while the pump laser is also slight asymmetric.

We have measured the REMPI signal of HD(v=1,j=0) as a function of difference frequency (detuning) and time-delay. The step-size are 0.01cm^{-1} and 1 ns, respectively. We got three dimensional surface from the data set after interpolation, as

showed in Figure S3. The plot shows two local maximums and a saddle point in between, in agreement with the SARP prediction.

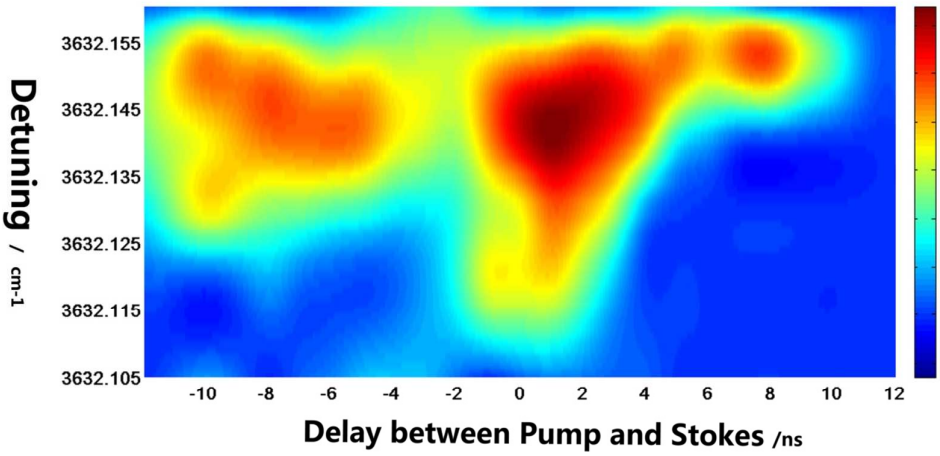

Figure S3. The REMPI signal of HD(v=1,j=0) as a function of detuning and time-delay.

Two fast photodiodes were used to measure the temporal jitter. The procedure is described as followed: one photodiode was used to detect the pump pulse and the generated electric signal was used to trigger the oscilloscope (Tektronix TDS5054). The energy of the pump pulse was very stable, so the oscilloscope was stably triggered. Another photodiode was used to detect the Stokes pulse, and the electric signal was displayed in the same oscilloscope. So we can directly read the temporal separation of two pulses. The oscilloscope ran in the "single shot" mode, and at each time the temporal separation was recorded. We repeated the "single shot" acquisition for 600 times, and then plotted the distribution of jitter. See Figure S4. The x-axis is the jitter (with 0.2 ns interval).The y-axis is the number of counts. Note the time-zero

of the jitter is determined artificially and have only relative meaning. The peak width (FWHM) of the Gaussian fit is 1.35ns.

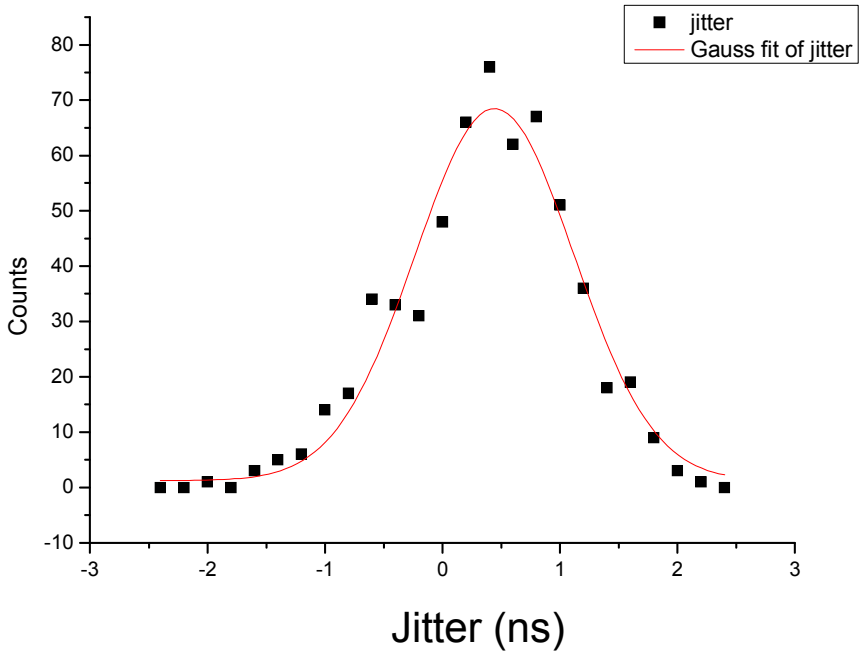

Figure S4. The distribution of the temporal jitter. Black dots are the experimental data points. The red line is the Gaussian fit.