High temperature source of pulsed supersonic beam of vdW complexes: from principle of operation to rotational structure in CdAr

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Molecular Spectroscopy and Quantum Information

Atomic Scale Science for Innovative Economy

Pulsed supersonic source

In molecular beam experiments supersonic pulsed sources are preferred over those operating in continuous mode. Unfortunately, commercially available pulsed valves can operate only up to 590K, which is insufficient for many purposes (particularly for production of supersonic beam of cadmium dimers). To overcome this limitation we use long plunger and water shields in conjunction with commercial solenoid valve (Parker-General Valve, Series 9). Implemented improvements have enabled construction of the source, which can operate up to 1000K and 10bar carrier gas stagnation pressure.

Construction

In the presented pulsed supersonic source, cadmium metal is heated in the lower reservoir (13) up to 950K and mixed with carrier Rg rare gas (Rg= Ar, Kr, Ne) delivered by a lower pipe (12). Next, the cadmium vapours (partial pressure 0.2bar) enter the upper source chamber (17) which ends with a nozzle (usually D=0.15mm orifice diameter, temperature 1000K) through which the mixture expands to the vacuum chamber forming supersonic beam that contains Cd_2 and CdRg van der Waals molecules. The details of construction are presented in Fig.1 (on the right) and also in [1].

Experiments, data acquisition and data analysis

In the experiment, molecules in supersonic beam were irradiated at a distances in the range from 8mm to 25 mm from the nozzle with a frequency-doubled dye laser beam. The excitation spectra were recorded in the 30690-30750 cm^{-1} spectral region by focusing the total laser induced fluorescence (LIF) from the interaction region on the cathode of a photomultiplier (PM) tube. During data acquisition phase, for each laser frequency the signal from PM was averaged in the oscilloscope (e.g. for 64 laser shots) and the resulting waveform was saved in a computer memory. The final spectrum can be determined from collected waveforms after selecting proper size of time-integration window. The selection of time-gating window have an impact on which type of molecules from supersonic beam (CdRg or Cd_2) contributed to the final spectrum, so different type of spectra can be obtained from one set of waveforms (see Fig.2 below).

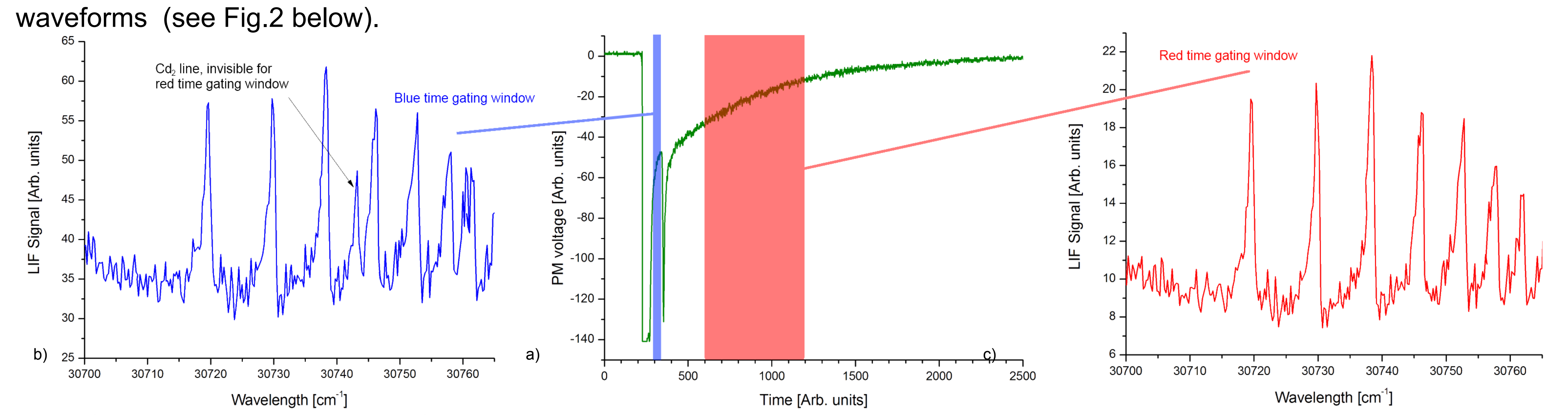

Fig. 2 a) Sample of the waveform (averaged signal from PM for one laser frequency), red and blue rectangles illustrate the time-gate interval applied for left and right spectra, respectively. b) LIF spectrum with Cd_2 and CdAr vibrational components determined with blue time gate window. c) LIF spectrum determined with red time-gate window. The Cd_2 vibrational components are now invisible, as the fluorescence from Cd_2 lives shorter than that from CdAr (i.e., Cd_2 fluoresces before opening of the integration window).

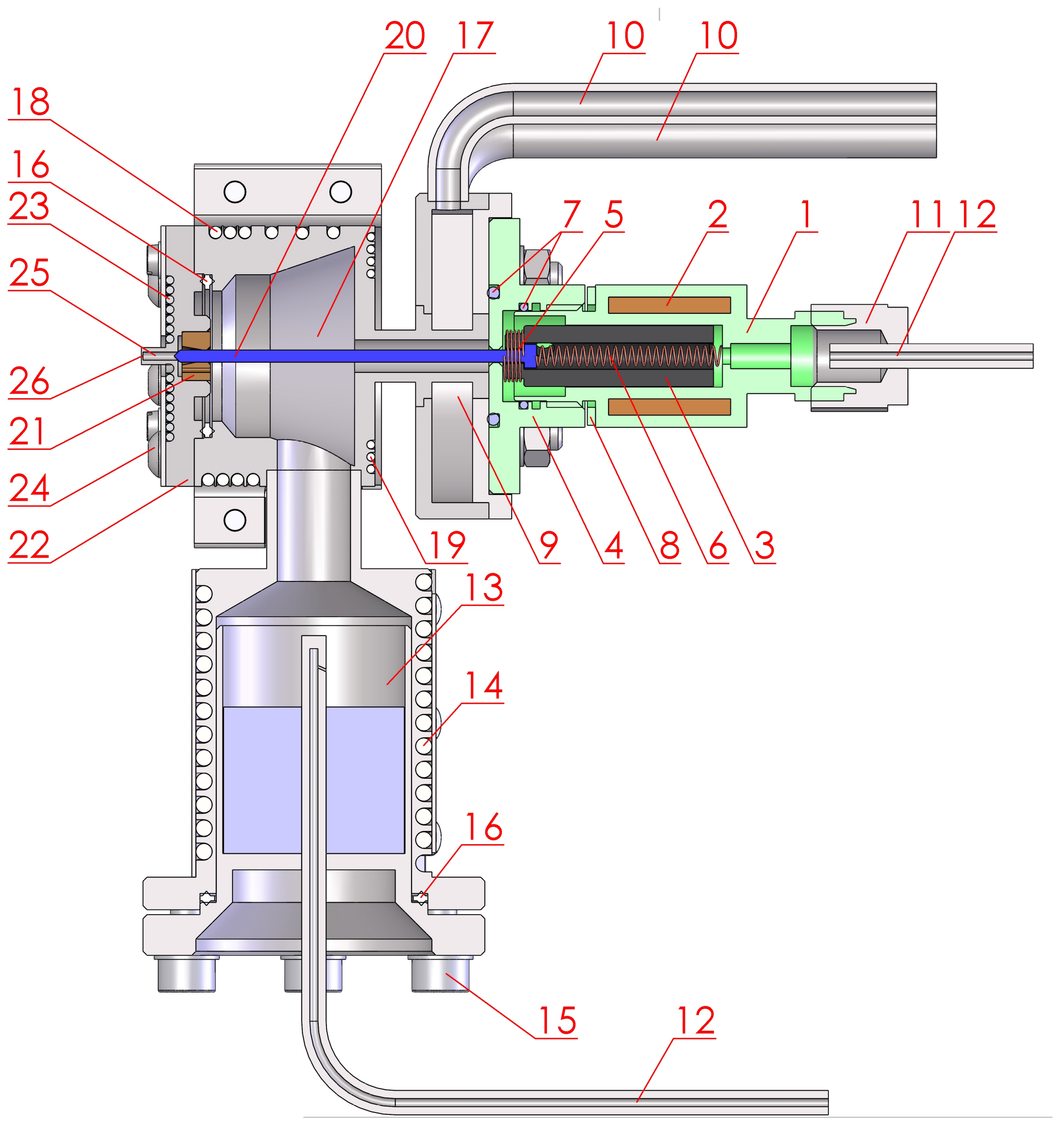

Fig.1. Cross-section of the high-temperature high-pressure pulsed source of vdW dimers developed in our laboratory. Parker General Valve (Series 9) commercial solenoid valve (green): 1, coil assembly; 2, solenoid coil; 3, armature; 4, flange mount; 5, buffer spring; 6, main spring; 7, Kalrez orings of DuPont™; 8, shim. Parts of the pulsed source: 9, water shield; 10, water pipes; 11, Swagelok® connection; 12, carrier gas supplies; 13, Cd reservoir; 14, reservoir heater; 15, reservoir screw; 16, iron washer; 17, source chamber; 18, source body heater; 19, source body back heater; 20, titanium plunger; 21, brass slide bearing; 22, nozzle cartridge; 23, nozzle cartridge heater; 24 nozzle cartridge screw; 25, nozzle channel; 26, orifice.

Analysis of partially resolved rotational structures in $B^31(5^3P_1) \leftarrow X^10^+(5^1S_0)$ transition in CdAr

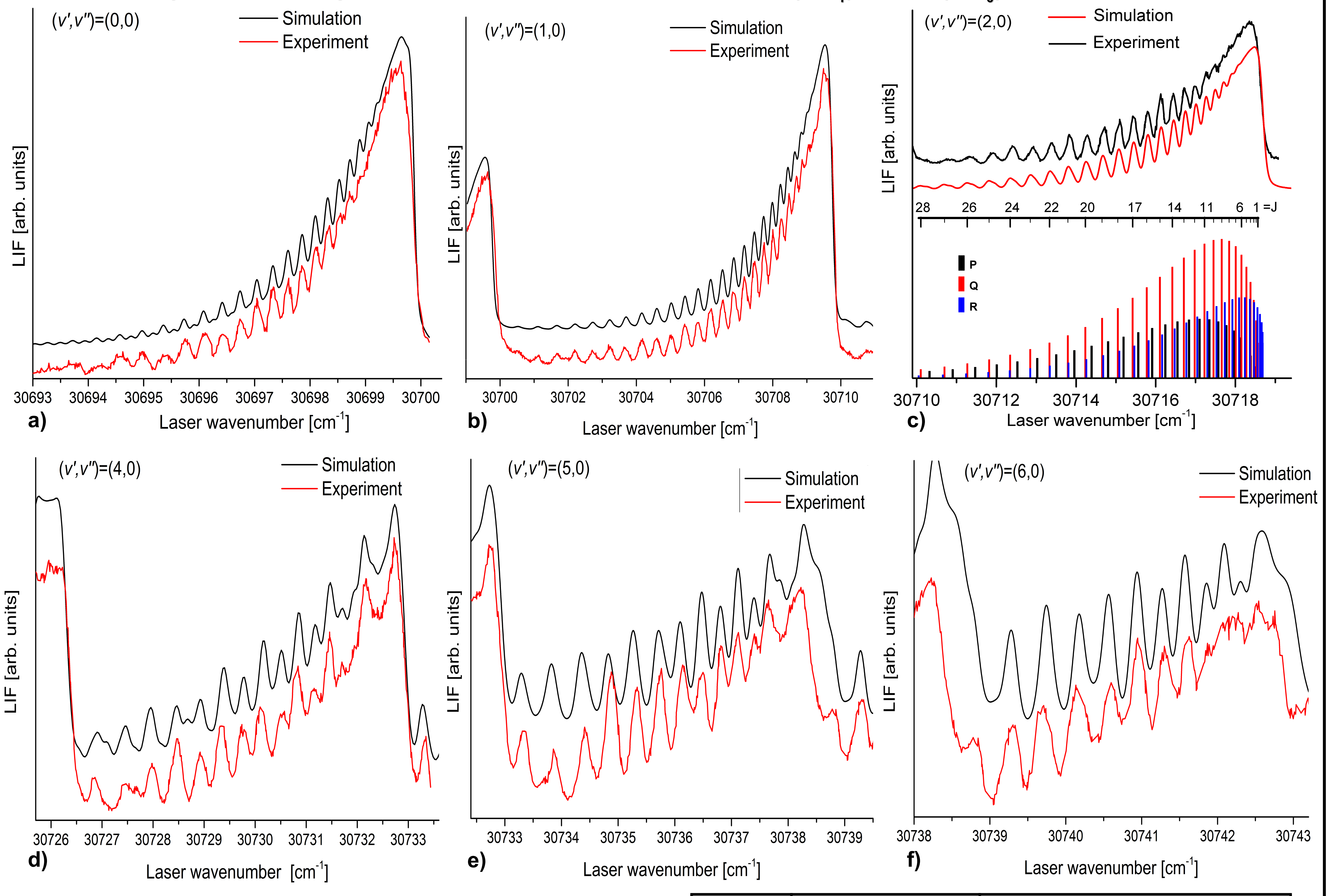

Fig. 3 a-f) Excitation spectra recorded (red) using the $B^31(5^3P_1) \leftarrow X^10^+(5^1S_0)$ transition in CdAr with respective PGOPHER [2] simulations (black). Each simulation includes all isotopologues and was performed for $B_{v'}$ and $D_{v'}$ values from Tab.1 (rotational constants for different isotopologue was recalculated considering differences between masses of isotopologues [3]). In the simulation, the rotational temperature T_R , laser bandwidth Δ_L and Gaussian broadening Δ_G were as follow: a) (7.5K, 0.06cm^{-1}, 0.11cm^{-1}), b) (7.1K, 0.06cm^{-1}, 0.15cm^{-1}), c) (8K, 0.06cm^{-1}, 0.15cm^{-1}), d) (9K, 0.06cm^{-1}, 0.12cm^{-1}), e) (8K, 0.06cm^{-1}, 0.15cm^{-1}), f) (8K, 0.06cm^{-1}, 0.15cm^{-1}).

v'	$B_{v'}$ [cm^{-1}]	$D_{v'}$ [cm^{-1}]
0	0.02215	4.0114e-7
1	0.02105	4.4367e-7
2	0.01994	4.8620e-7
4	0.01735	6.1289e-7
5	0.01616	6.6467e-7
6	0.01497	7.1645e-7

Tab.1. Rotational constants of the B^31-state vibrational bands for the most abundant $^{114}Cd^{40}Ar$ isotopologue.

Control over rotational temperature

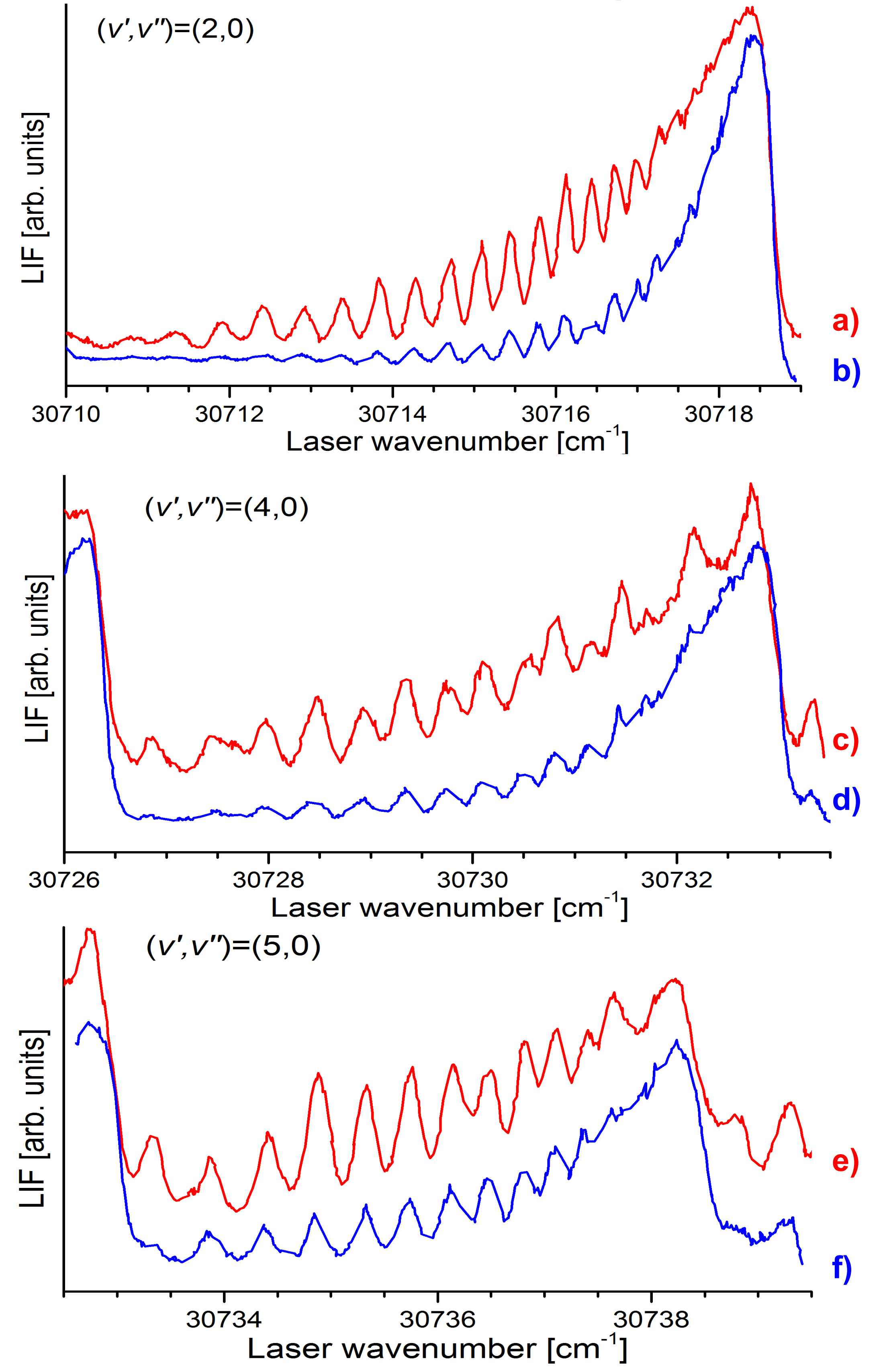

Fig.3. Proper choice of parameters of expansion in the supersonic beam (e.g., nozzle diameter, temperature of the source and carrier gas pressure) influences T_R rotational temperature and can significantly simplify the data analysis. It can be seen, that for low v' higher T_R is preferable due to the excitation of states with higher J', although for higher v' the optimum T_R is lower due to an overlaps of the neighbouring bands of different v'. The measured T_R is: a) 8K, b) 4.5K, c) 9K, d) 4.3K e) 8K, f) 5K. Spectra a), c) and e) collected for 0.25 mm nozzle diameter (red), spectra b), d) and f) collected for 0.18 mm nozzle diameter (blue).

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References

[1] T. Urbanczyk, J. Koperski, *Rev. Sci. Instrum.* **83**, 083114 (2012).
[2] PGOPHER, a Program for Simulating Rotational Structure, C. M. Western, University of Bristol, http://pgopher.chm.bris.ac.uk.
[3] T. Urbanczyk, J. Koperski „Pulsed supersonic source of vdW complexes for high-temperature applications: Spectroscopy and beam characteristics", *Eur. Phys. J. - Special Topics* (2013), in print.