SAND2021-15140C

PRINCETON UNIVERSITY

Rotation-vibration non-equilibrium measurement using pure rotational fs/ps CARS coherence beating

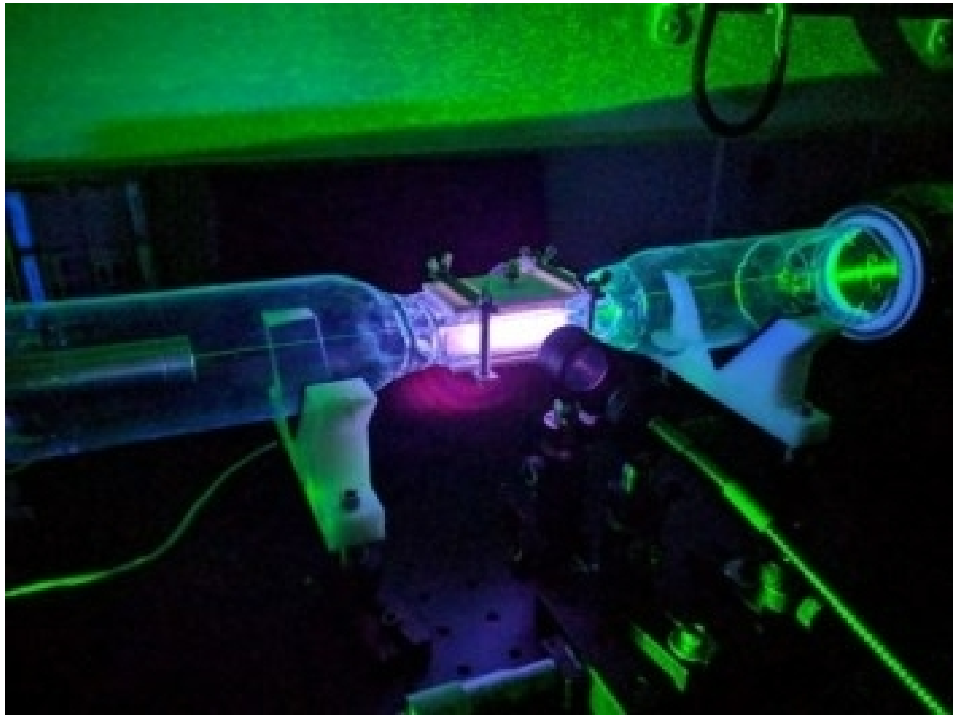
TIMOTHY Y. CHEN[1*], NING LIU[1],
ARTHUR DOGARIU[1], EGEMEN KOLEMEN[1,2], AND YIGUANG JU[1]

AIAA SCITECH FORUM, JANUARY 3-7, 2022 (PDL-09)

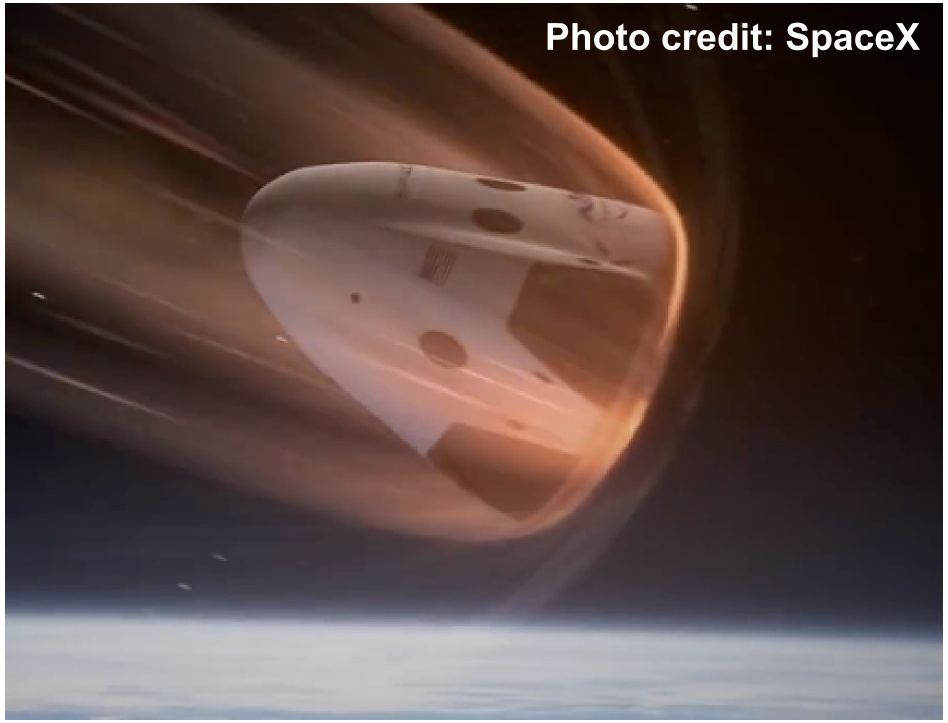
[1] *Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544*
[2] *Princeton Plasma Physics Laboratory, Princeton, New Jersey, 08540, USA*
**Presently at Sandia National Laboratories, Livermore, CA*

Rotation-vibration non-equilibrium is important for plasmas and hypersonics

$$T_{rot} \neq T_{vib}$$

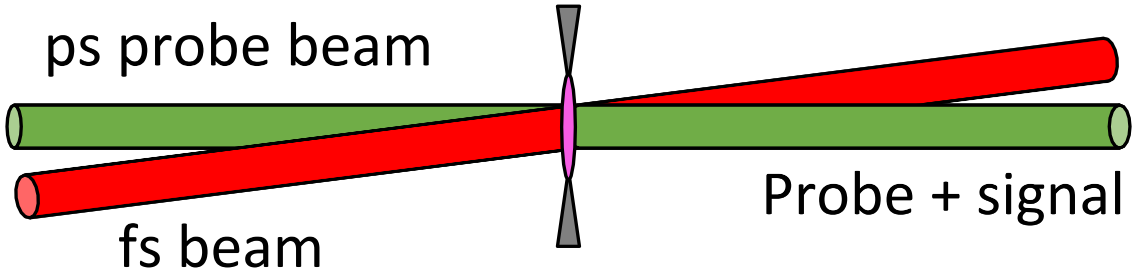


Chemical reactivity and thermalization

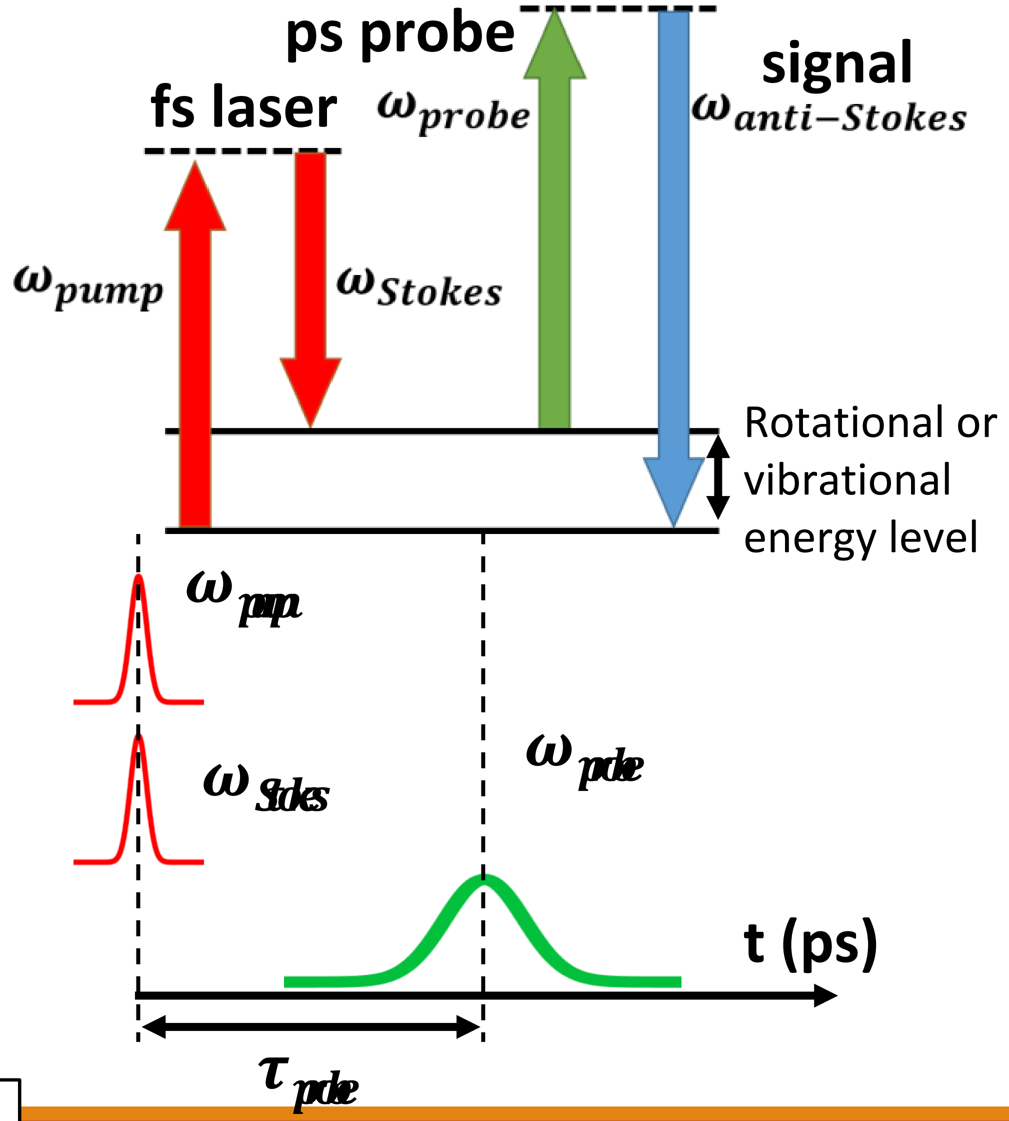


Hypersonic vehicle thermal loading and aerodynamics

What is hybrid fs/ps CARS?



- Use broadband femtosecond pulse for simultaneous acquisition of many Raman transitions and picosecond probe for spectral resolution [1-2]
- Several advantages:
  - $I_{CARS} \propto I_{fs}^2 * I_{ps} \rightarrow$ 1-D or 2-D imaging of temperature and species concentrations within 50 μm of surface [3-5]
  - No need to scan Stokes frequency and only needs 2 beams



[1] Pestov et al., Science (2007), [2] Prince et al. J. Chem Phys (2006), [3] Bohlin et al. J. Chem. Phys (2013), [4] Retter et al. Combust. Flame (2018) [5] Bohlin et al. Proc. Combust. Inst. (2017)

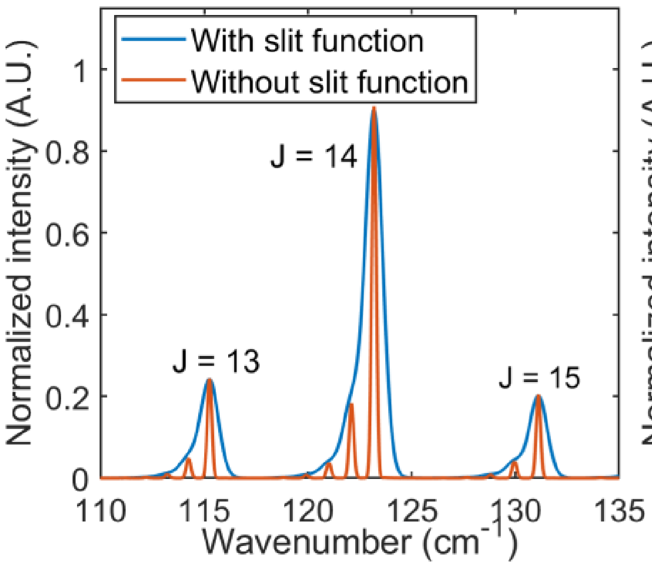
We can use pure rotational fs/ps CARS for simultaneous T_{rot} and T_{vib} measurements

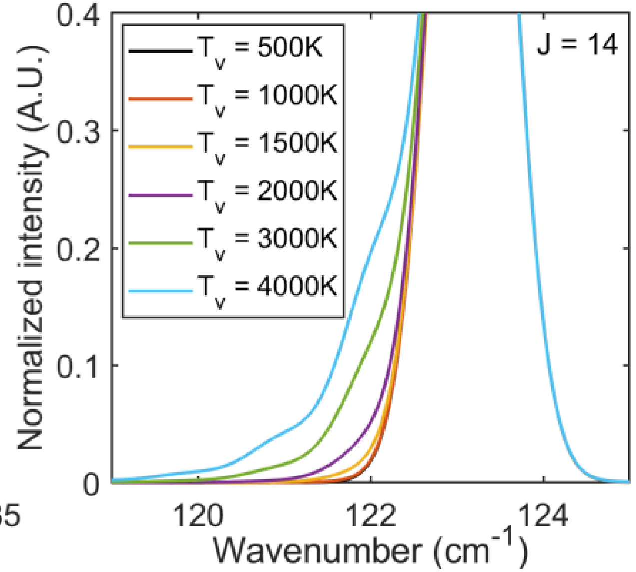
Rotational energy level $F(v,J)$

$$F(v,J) = B_v\big(J(J+1)\big) - D_v\big(J^2(J+1)^2\big)$$

$$B_v = B_e - \alpha\left(v+\frac{1}{2}\right) + \gamma_e\left(v+\frac{1}{2}\right)^2$$

v = vibrational quantum number
J = rotational quantum number
B_v = rotational constant
α = rotation-vibration coupling constant
D_v = centrifugal constant





Chen T.Y. et al. Opt. Lett. *(2020)*

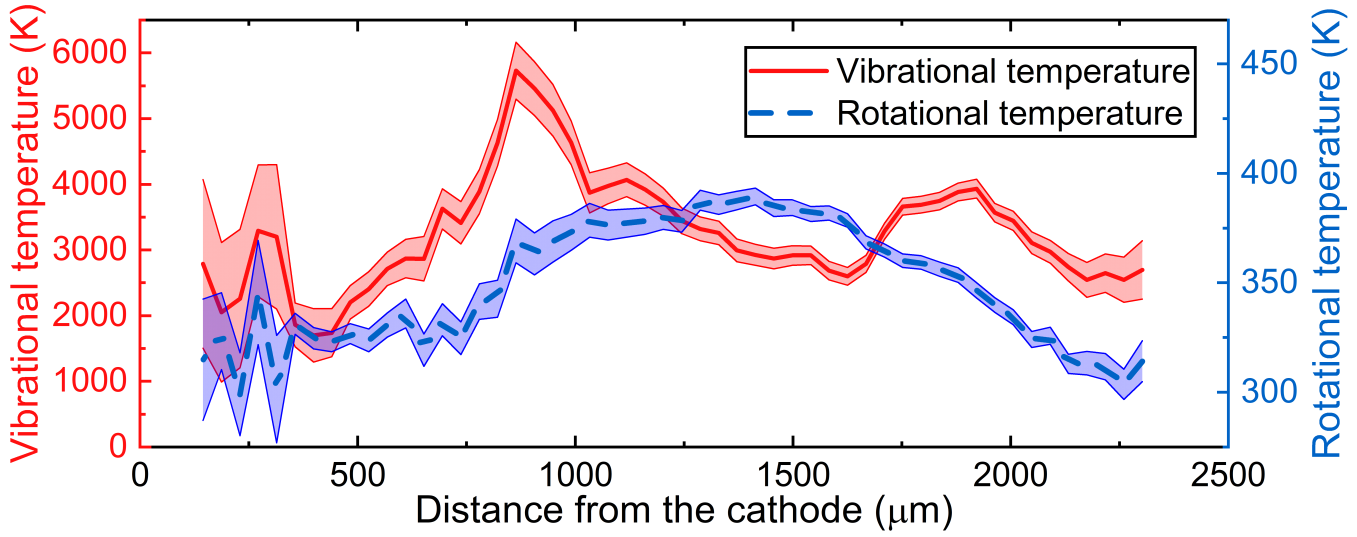
We can use pure rotational fs/ps CARS for simultaneous T_{rot} and T_{vib} measurements

Rotational energy level $F(v,J)$

$$F(v,J) = B_v(J(J+1)) - D_v(J^2(J+1)^2)$$

$$B_v = B_e - \alpha\left(v+\tfrac{1}{2}\right) + \gamma_e\left(v+\tfrac{1}{2}\right)^2$$

v = vibrational quantum number
J = rotational quantum number
B_v = rotational constant
α = rotation-vibration coupling constant
D_v = centrifugal constant



Demonstrated 1-D imaging of rotation-vibration non-equilibrium

Chen T.Y. et al. Opt. Lett. *(2020)*

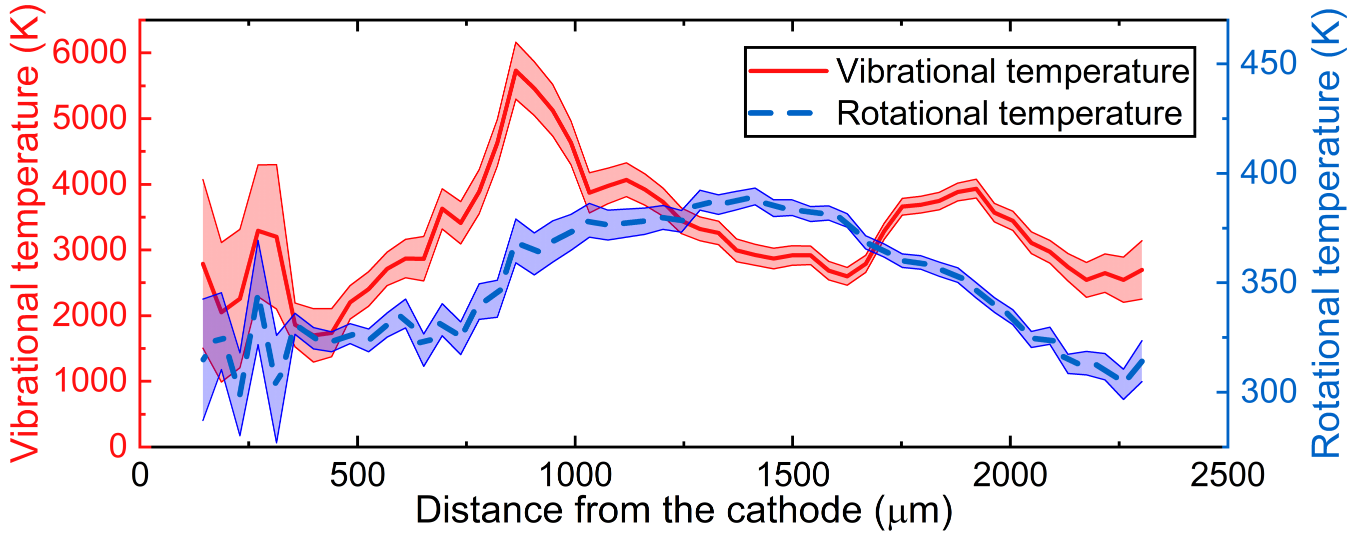
We can use pure rotational fs/ps CARS for simultaneous T_{rot} and T_{vib} measurements

Rotational energy level $F(v,J)$

$$F(v,J) = B_v\big(J(J+1)\big) - D_v\big(J^2(J+1)^2\big)$$

$$B_v = B_e - \alpha\left(v+\tfrac{1}{2}\right) + \gamma_e\left(v+\tfrac{1}{2}\right)^2$$

v = vibrational quantum number
J = rotational quantum number
B_v = rotational constant
α = rotation-vibration coupling constant
D_v = centrifugal constant

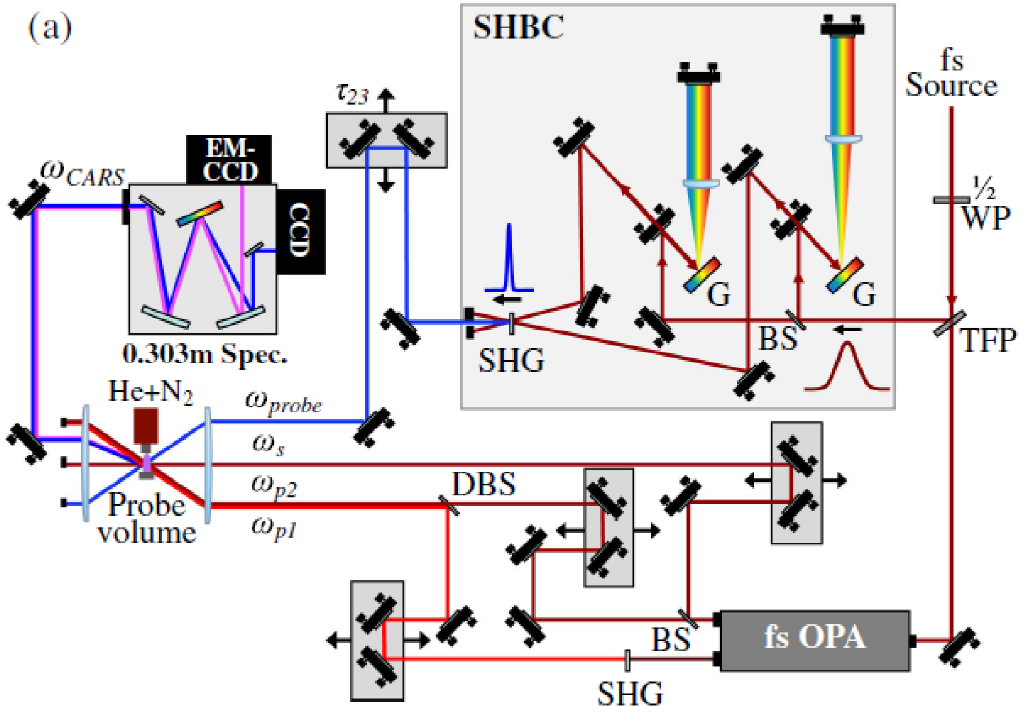


Needs a spectrally narrow laser ($< 0.5\ cm^{-1}$)

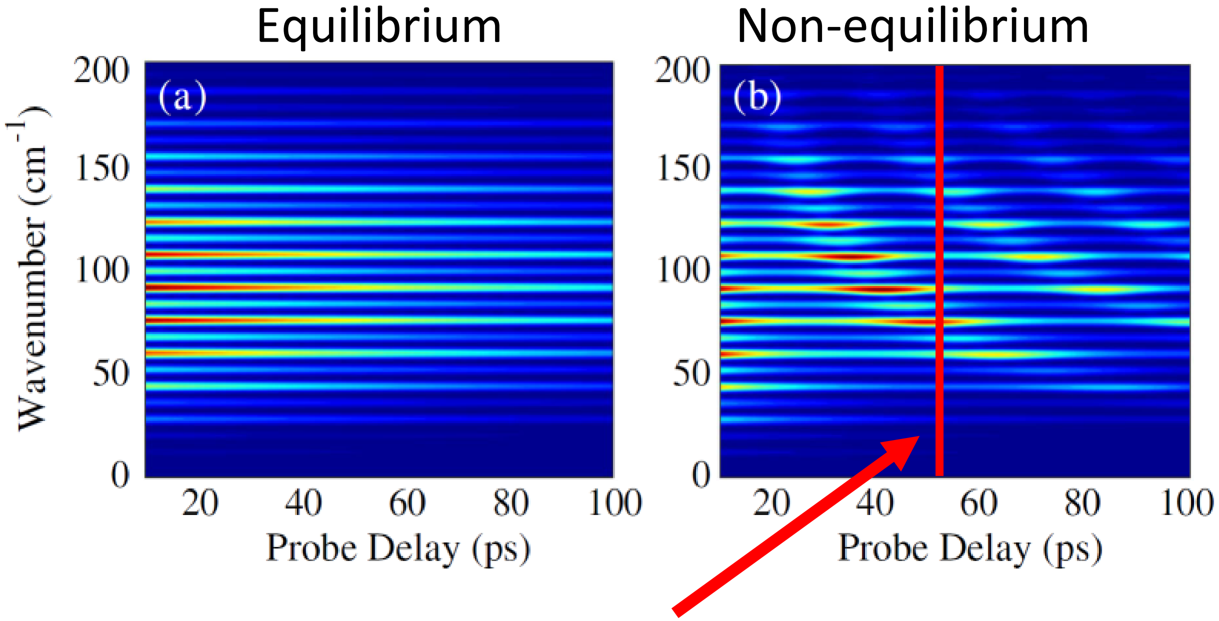
Chen T.Y. et al. Opt. Lett. *(2020)*

Is there a simpler method?

Dual pump fs/ps CARS is sensitive uses one fs laser but needs **4 beams**

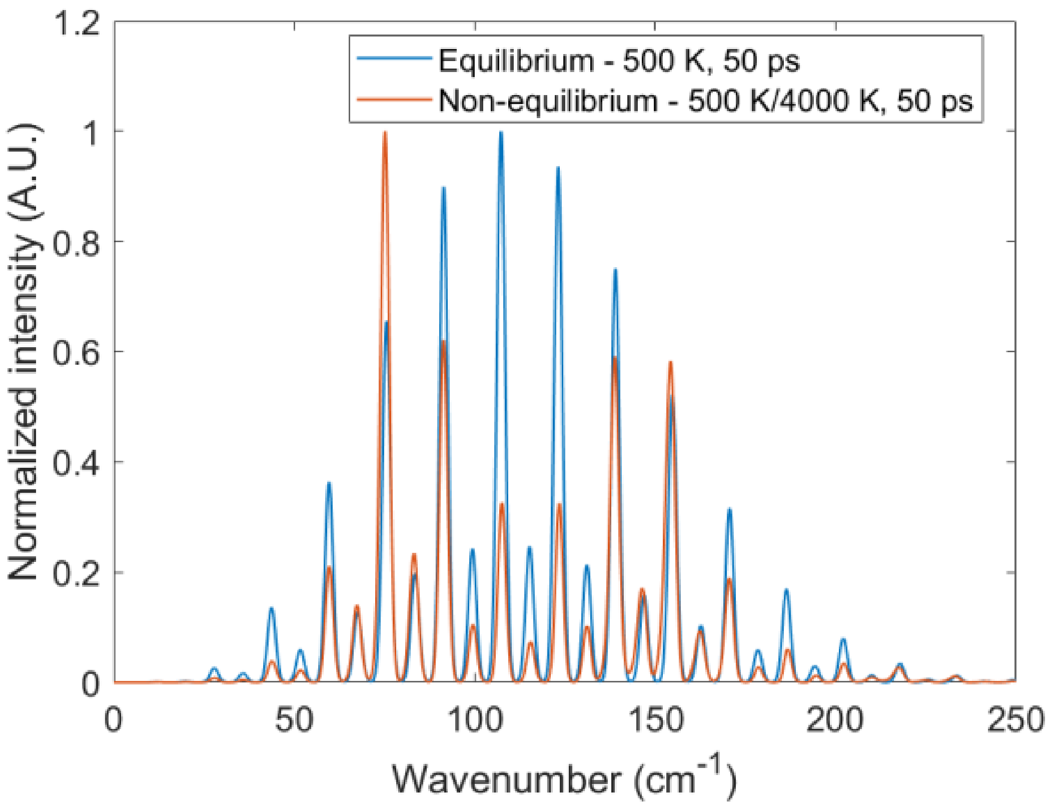


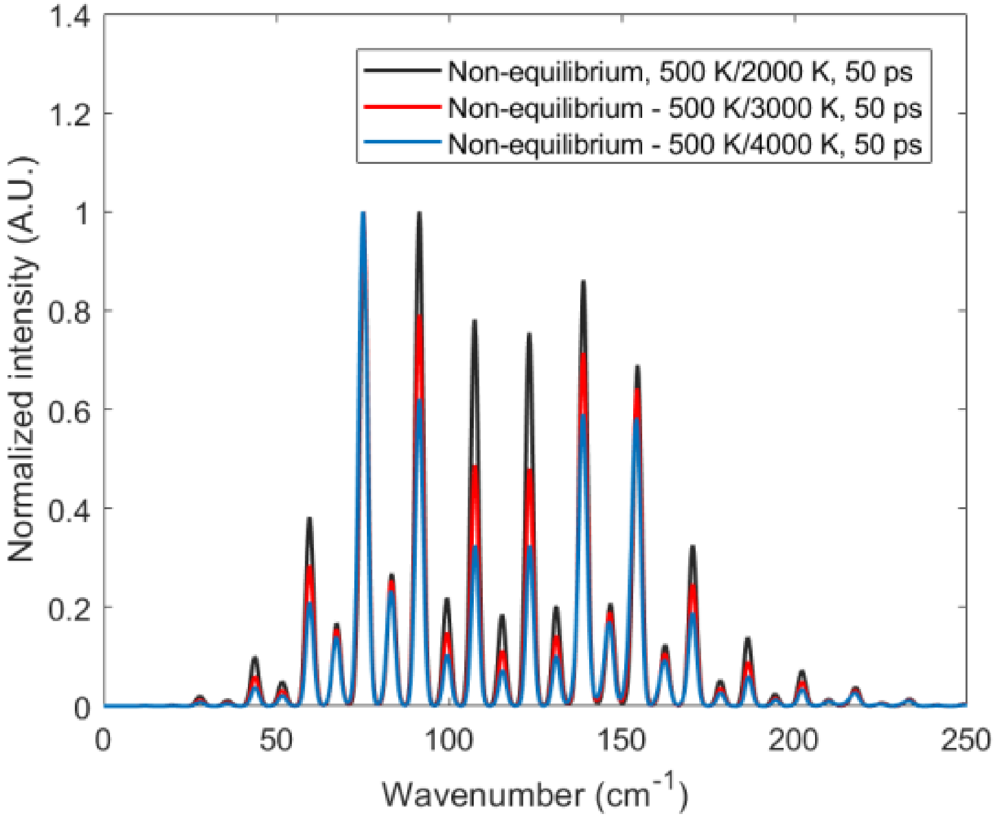
Dedic, C.E. et al., *Optica*, (2017)



If we know probe delay and beating pattern, can we extract T_{vib} and T_{rot} ?

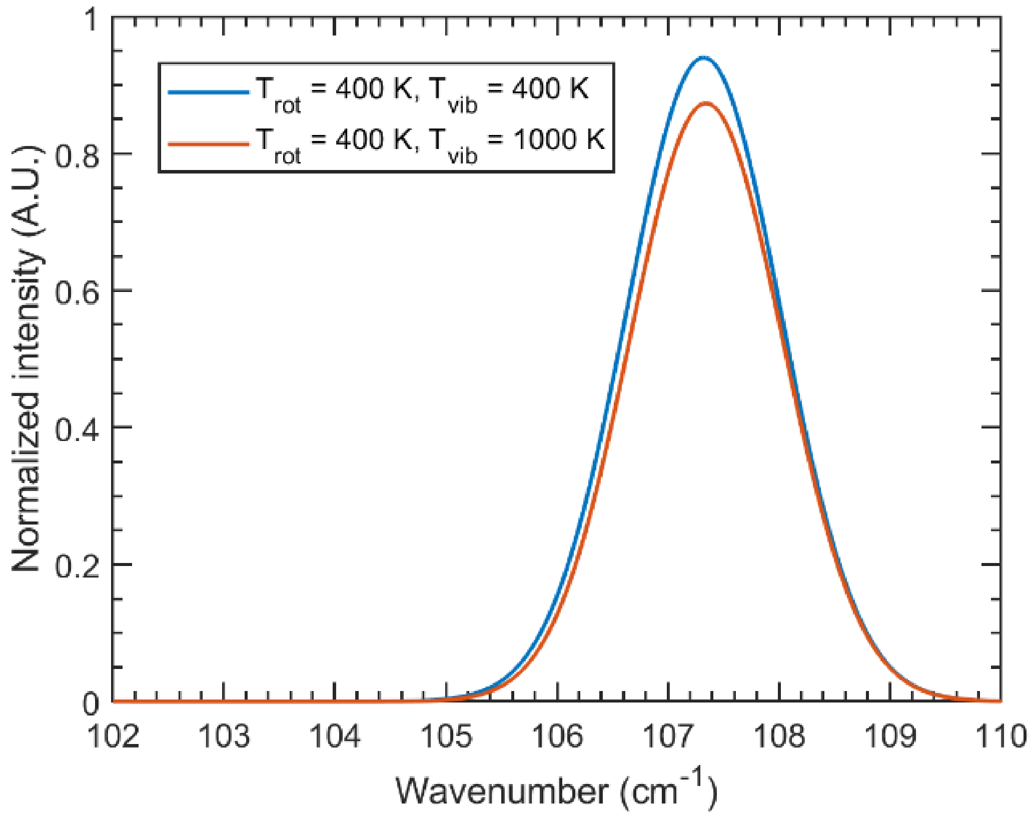
We can use coherence beating for simultaneous T_{rot} and T_{vib} measurements



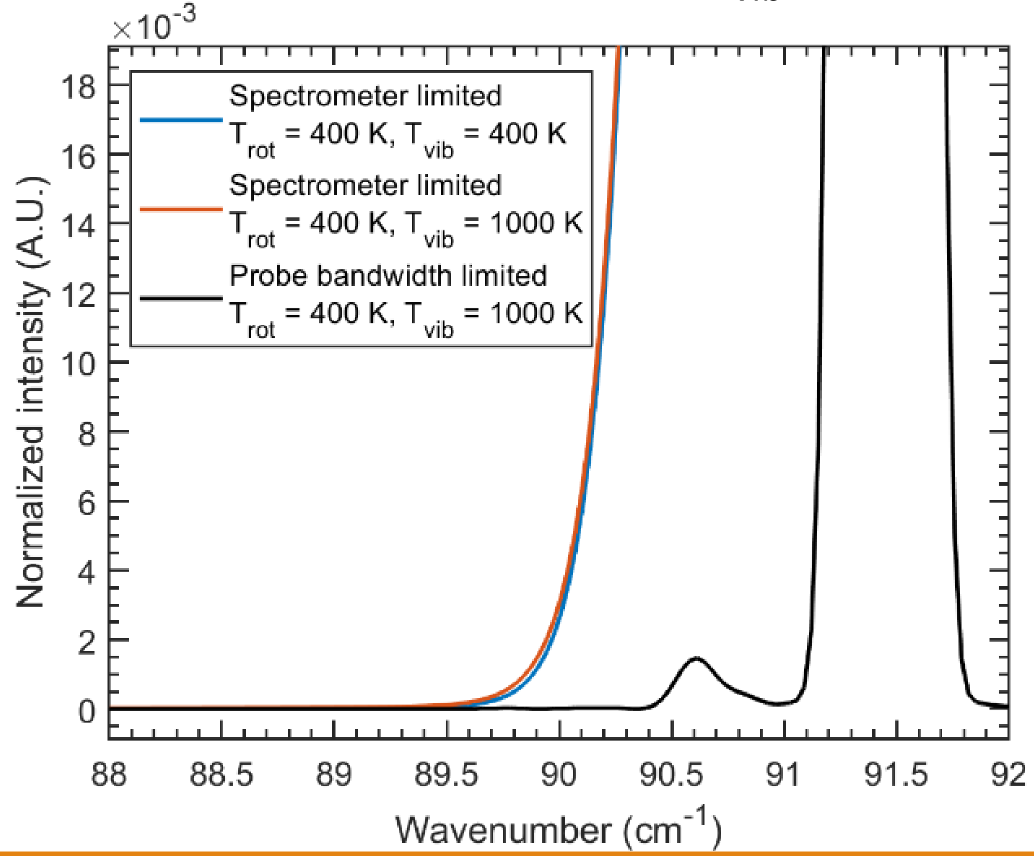


How does the sensitivity compare to frequency domain detection?

Sensitive to T_{vib} = 1000 K

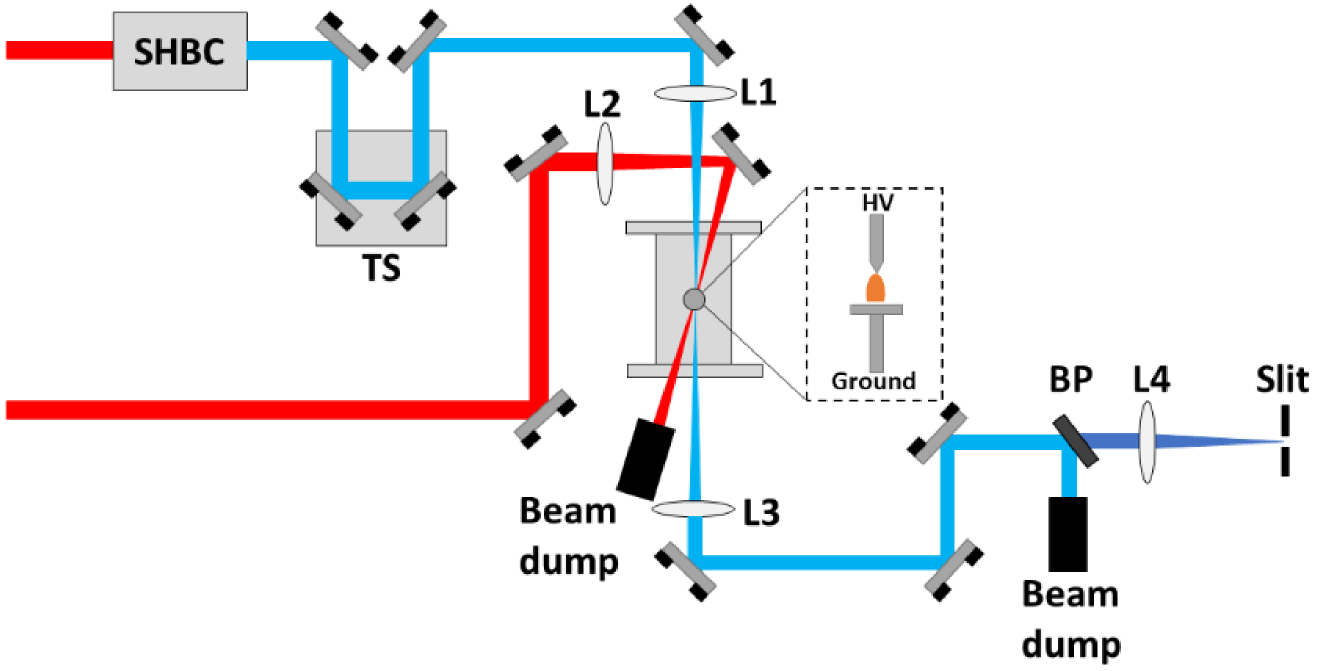


High spectrometer resolution needed for measurement of T_{vib} = 1000 K



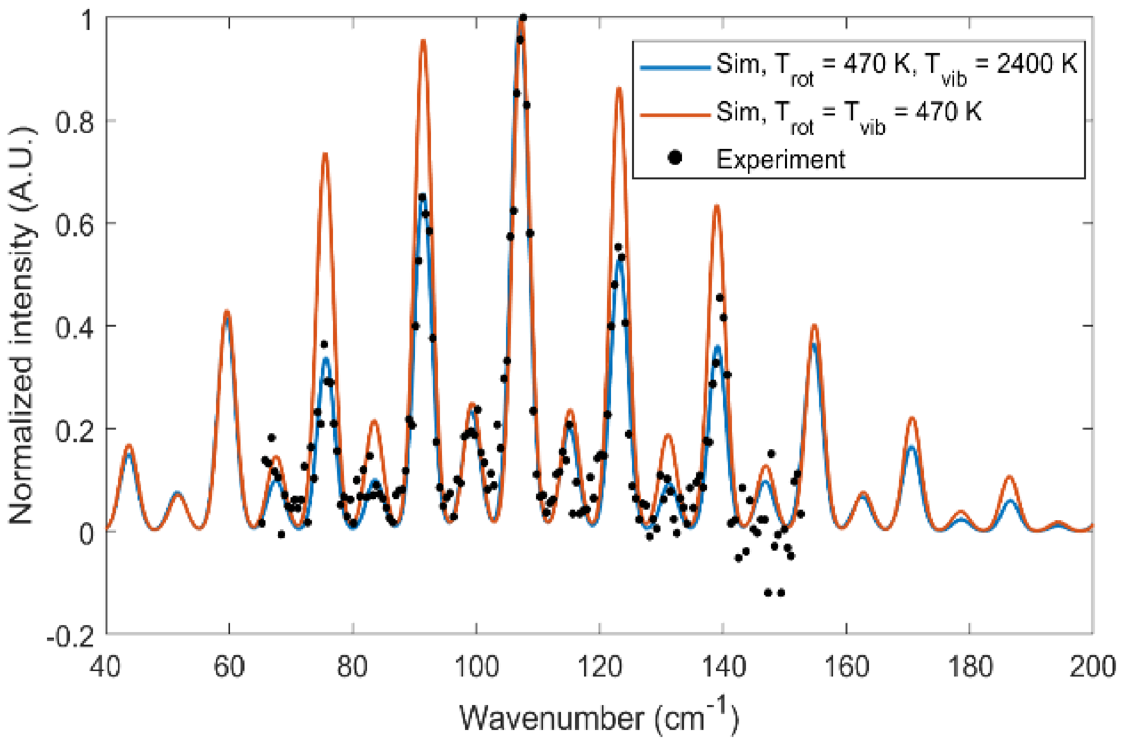
Experimental setup for model validation

75 Torr pure N_2, 2 kV DC, 100 kΩ resistor in series, I ~ 15 mA

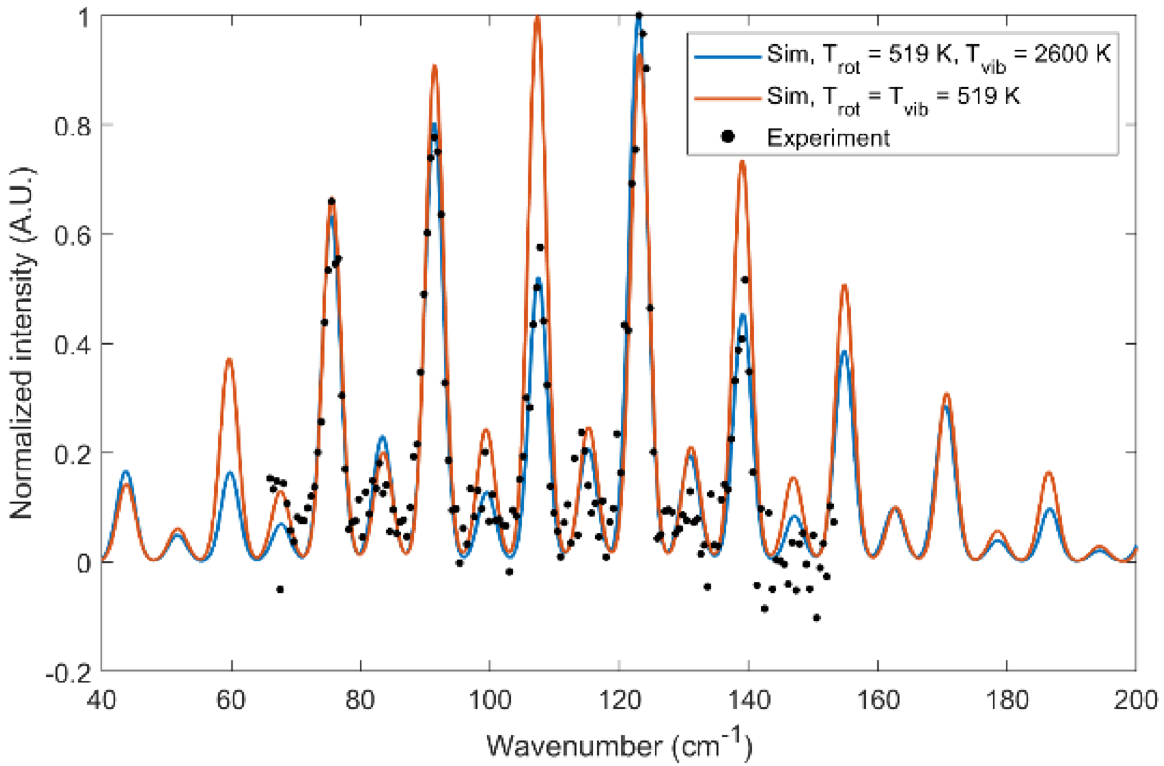


Simulated spectra fit well with the measurements

T_d = 72 ps

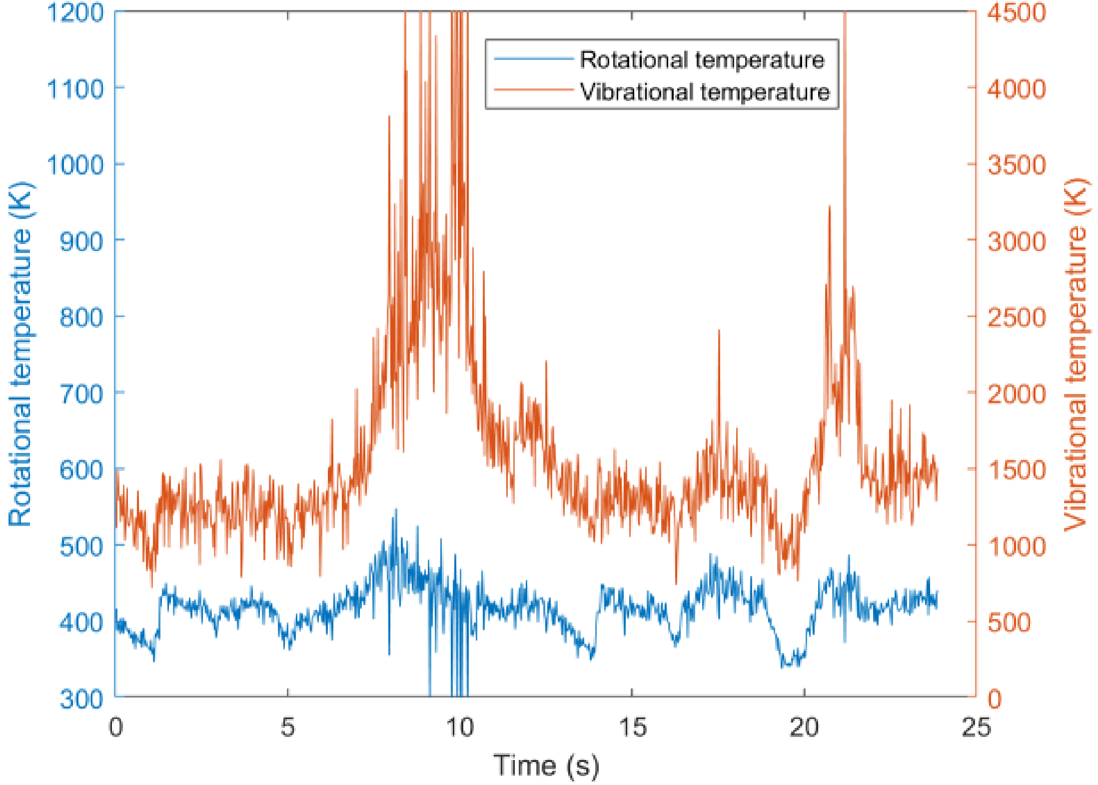


T_d = 92 ps

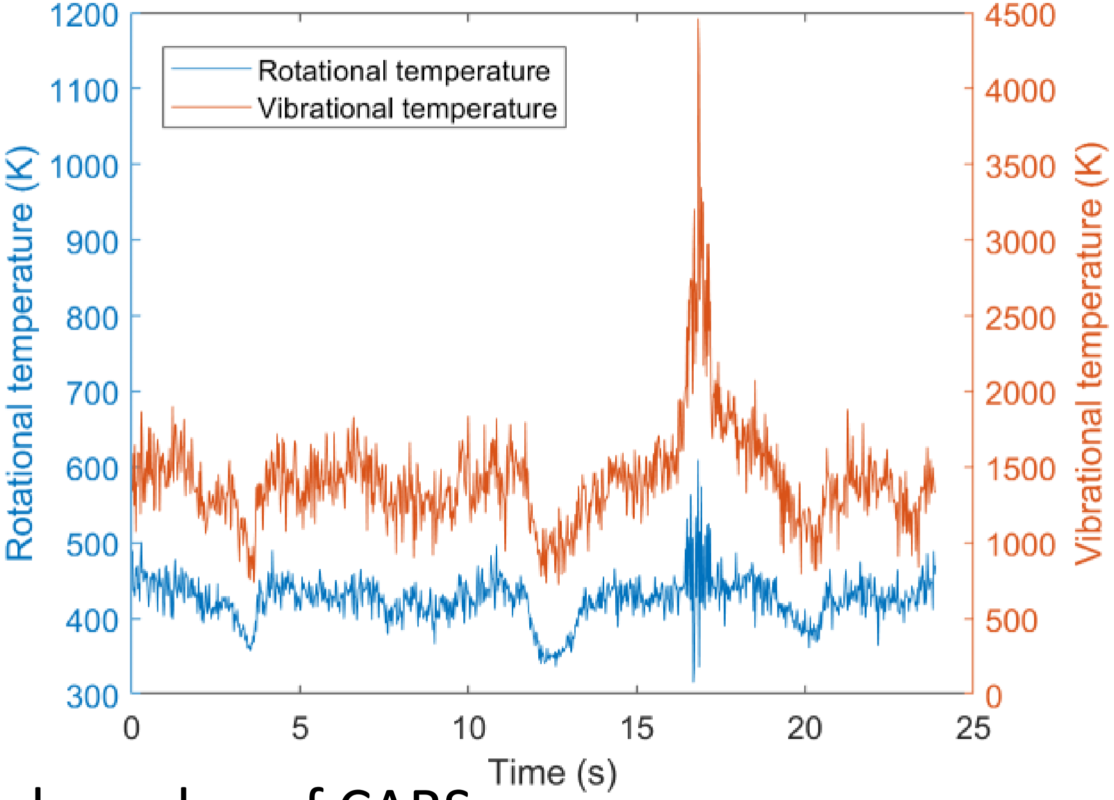


Time traces at two probe delays show consistent temperatures and dynamics

t_{delay} = 72 ps



t_{delay} = 92 ps



Peaks are likely optimal overlap of CARS probe volume and positive column of plasma

Conclusions

- Developed new diagnostic for simultaneous T_{rot} and T_{vib} measurement using pure rotational fs/ps CARS coherence beating
- Can be more sensitive than detection in frequency-domain
- Demonstrated this technique with a N_2 DC discharge and successfully fit and extracted non-equilibrium T_{rot} and T_{vib}
- Enables high speed 1-D and 2-D CARS measurements of rotation-vibration non-equilibrium in plasmas and hypersonic flows

Acknowledgements

This work was supported by ExxonMobil through its participation in the Princeton E-filliates Partnership of the Andlinger Center for Energy and the Environment. TC was partially supported through the Program in Plasma Science and Technology at Princeton University (PPST) Fellowship.

Questions?