INSTITUT DE CHIMIE
SEPARATIVE DE MARCOULE

# Sonoluminescence Spectra Generated in Aqueous Solutions by a 4-MHz HIFU in Vobulated Mode

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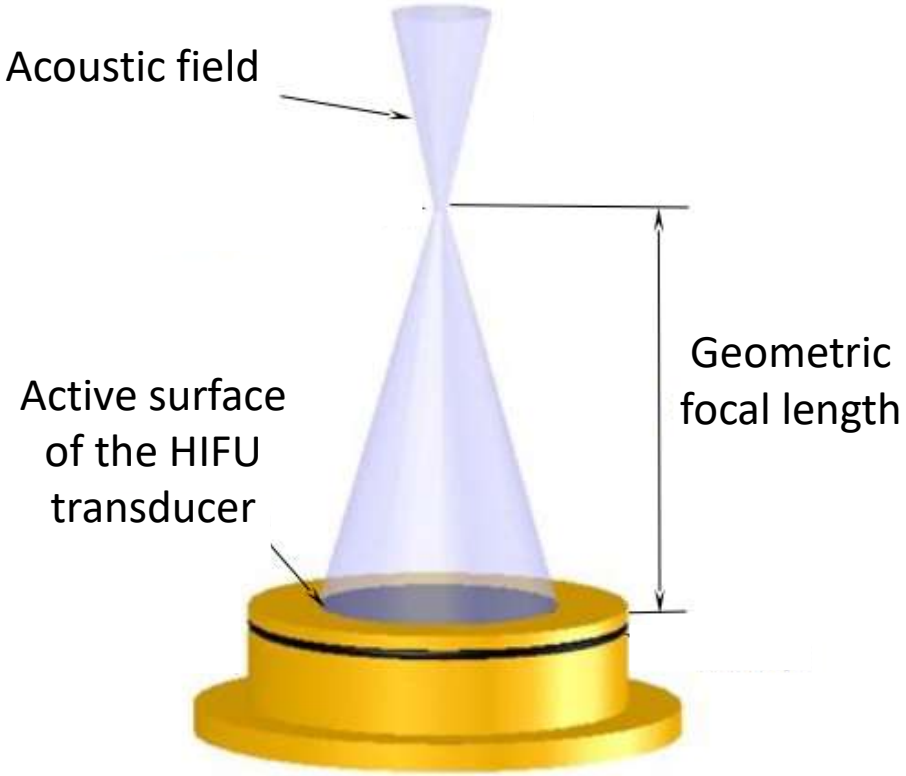
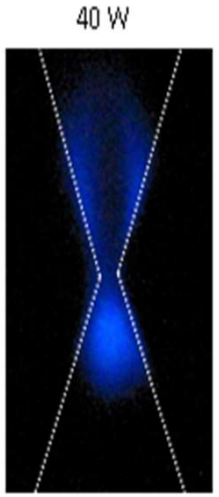
# HIFU: High Intensity Focused US

HIFU: focus the US wave to increase the acoustic power density at one point

At very high frequencies, focusing US is the only possibility to exceed the cavitation threshold



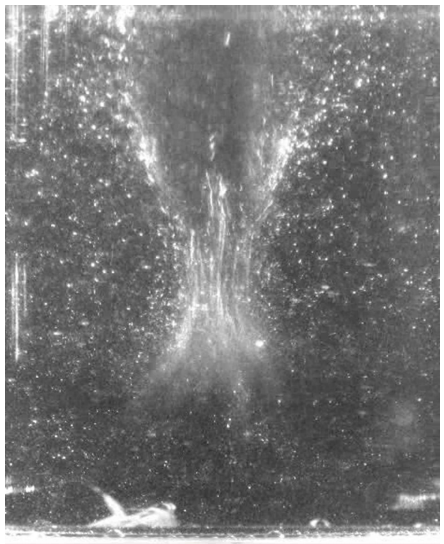

Luminol
sonochemiluminescence
(SCL)

# Tuning cavitation effects

## Sonochemistry and Surface Reactivity

### Sonochemical applications

Oxidation of chemical species



***Need maximum of inertial cavitation***

### Surface irradiation

Selective ablation of surface coating

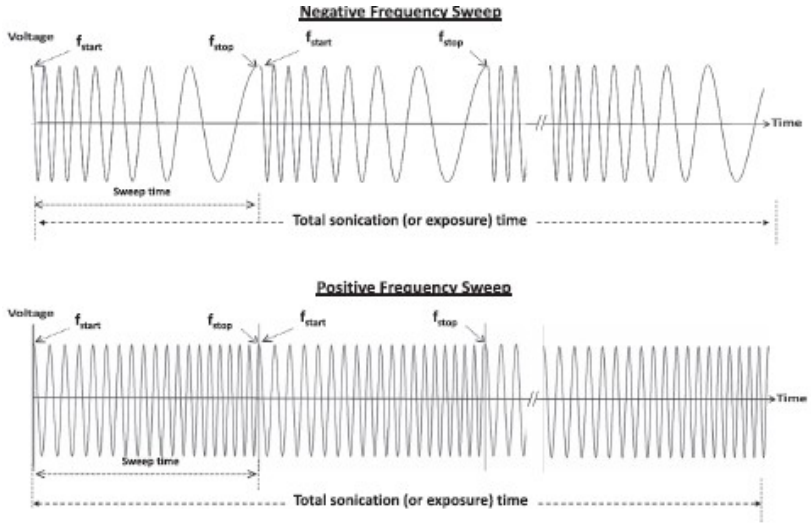
**Creep of melted polymer:** *Heat absorption*

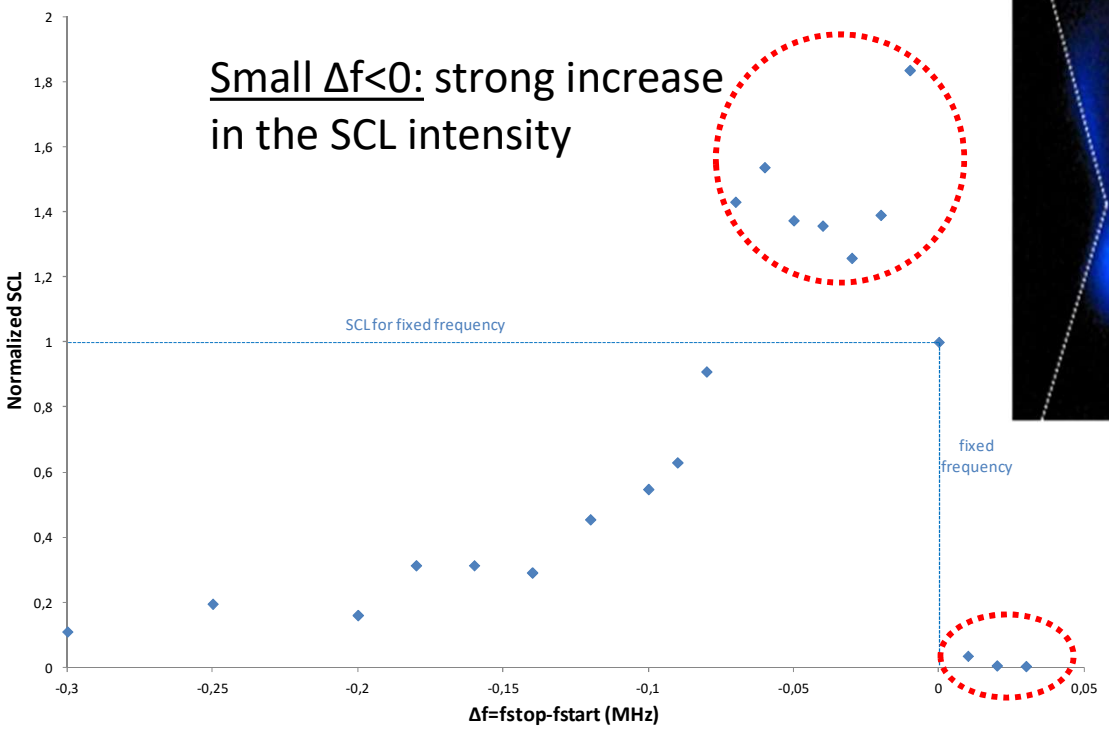

***Need to avoid cavitation***

→ Control operating parameters to get the expected cavitation effects

# Vobulation & SCL intensity

Vobulation = sweep in frequency





Attributed to changes in the number of active bubbles

$f_0 \rightarrow f_0 - |\Delta f|$

$R_0 \rightarrow R$ with $R > R_0$

Bubbles can grow during the pulse and become active

Δf>0: quenching of SCL

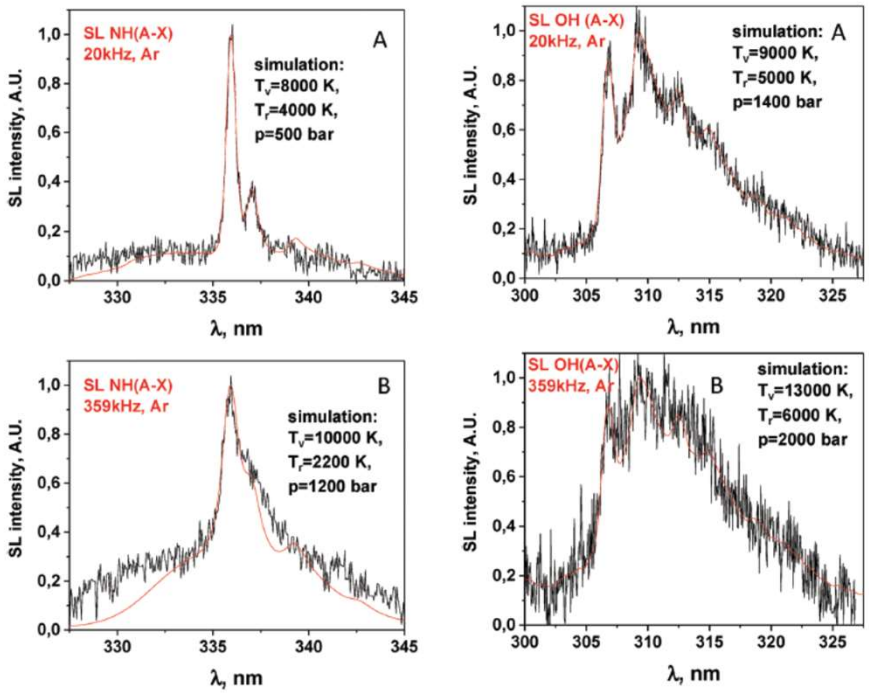
Can SL spectroscopy confirm this interpretation, and give some information on the conditions reached inside collapsing bubbles?

# Measuring HIFU SL spectra

## Very dim SL intensity

- Strong decrease in the SL intensity above ≈ 500 kHz
- Small zone of SL emission

Strong **broadening of molecular emissions** with increasing frequency: will they still be visible and will fitting be feasible?



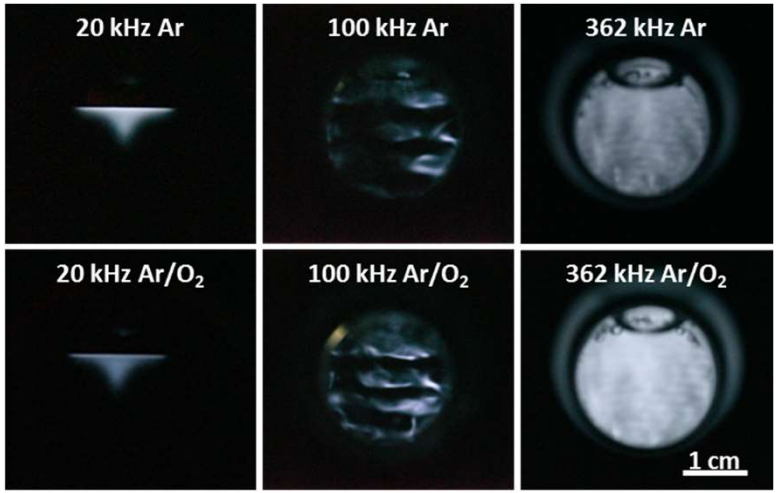

0.1M ammonia, Ar
20 kHz vs. 359 kHz

# HIFU SL spectra under Ar-20%$O_2$

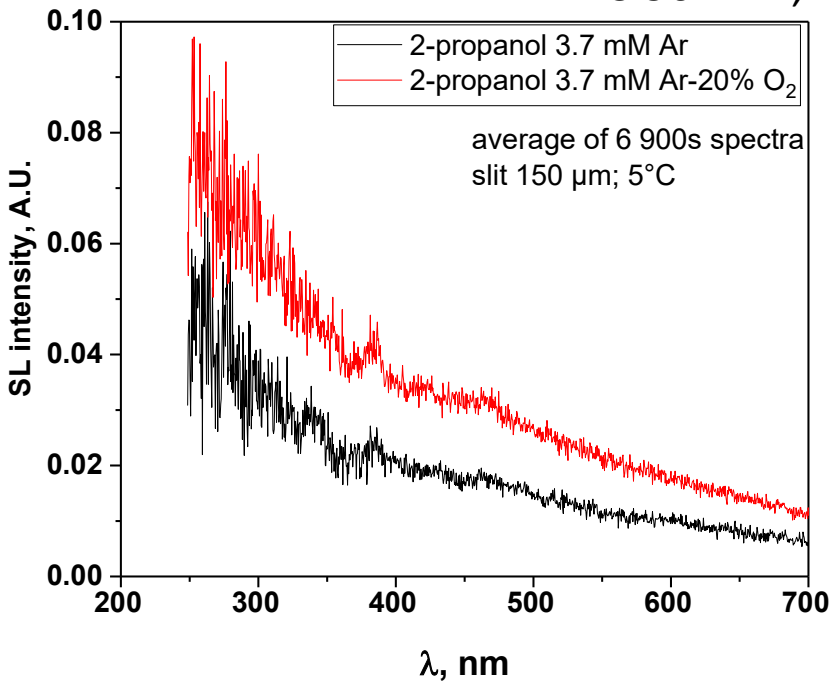
**Very dim SL intensity:**

**Use of Ar-20%$O_2$ as a saturating gas**

known to increase the SL intensity at high frequency due to $O_2$ dissociation




4 - 3.96 MHz; 4 ms



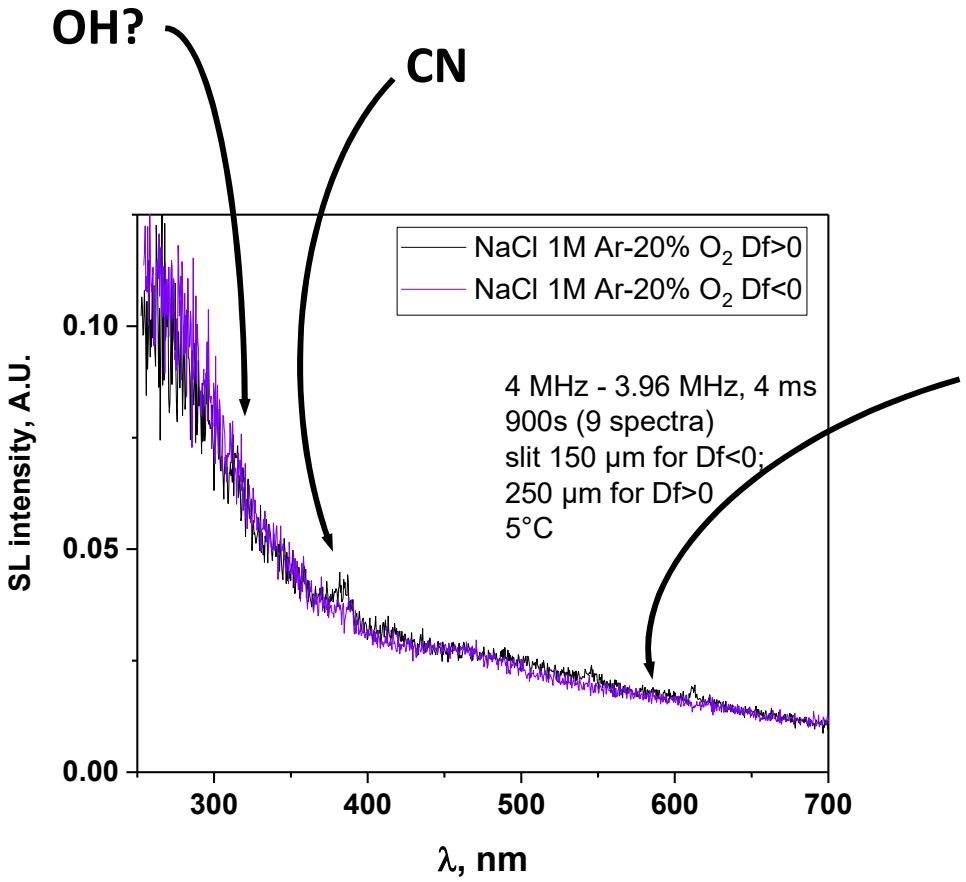

Ar-20%$O_2$: increase in the SL intensity

**$O_2$ dissociation happens at 4 MHz HIFU**


Pflieger et al., *Ultrason. Sonochem.* (2015)

Ji et al., *J Phys Chem C* 2018

# NaCl 1M Ar-20%$O_2$ SL spectra

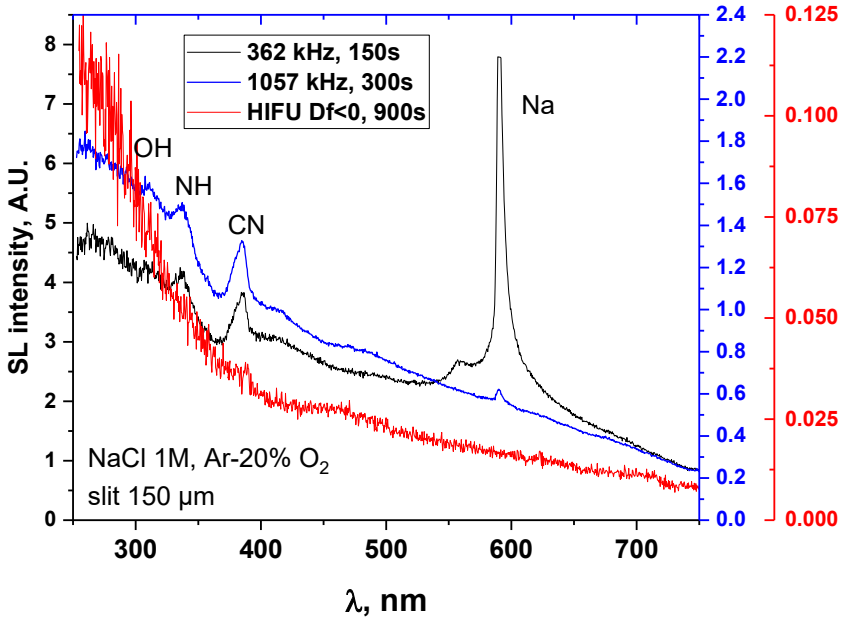



HIFU SL continuum intensity (at 500 nm):

- 100 times lower than at 1057 kHz
- 600 times lower than at 362 kHz

**No Na* emission**

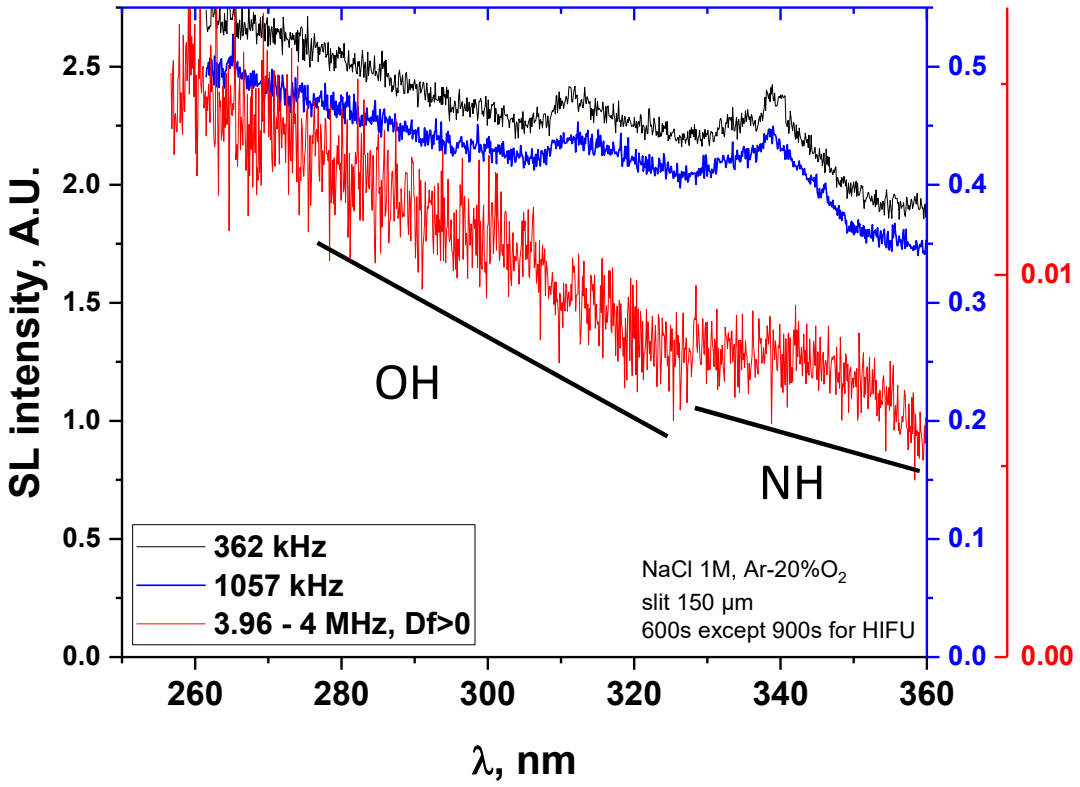
Strong decrease of Na emission intensity with US frequency
(smaller bubbles & less droplet injection)




**HIFU: very different continuum shape!**

# What about molecular emissions?

NaCl 1 M, Ar-20%$O_2$
Higher resolution UV grating (0.83 nm)
4 - 3.96 MHz; 4 ms




**Very broad** not well defined molecular emissions (OH & NH)

Presence of NH due to traces of air:

$N_2$ ))) → N + N

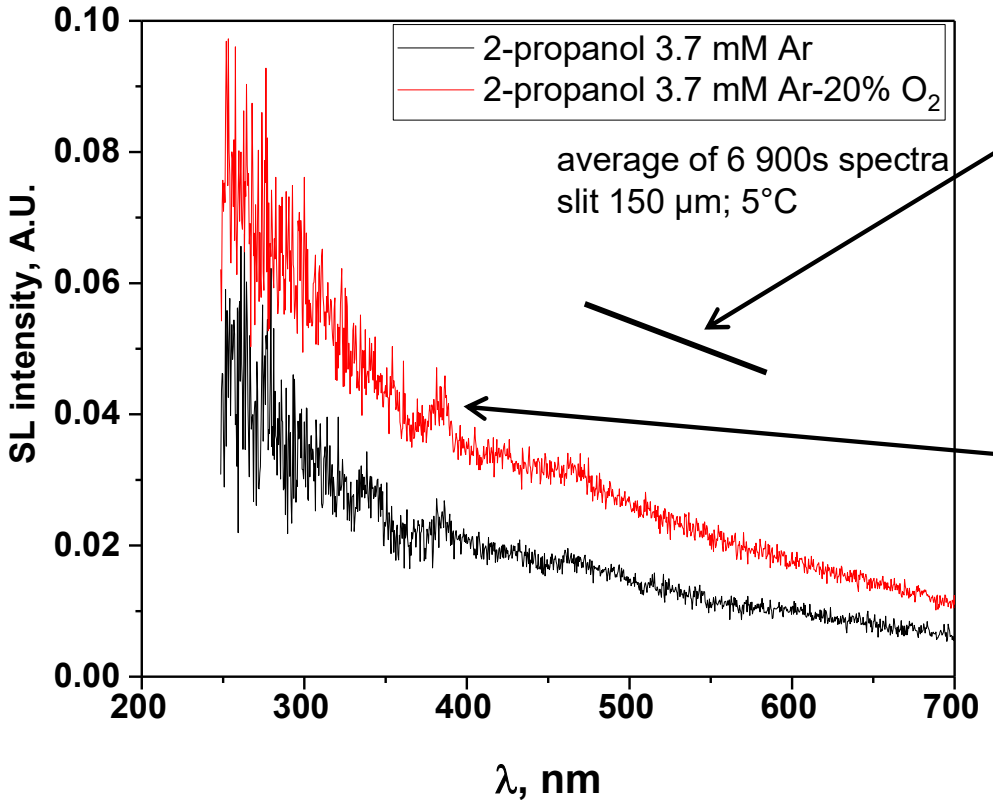
N + $H_2$ → NH + H

N + H → NH

**$N_2$ dissociation occurs in 4 MHz cavitation bubbles**

# Other known molecular emissions?

2-propanol 3.7 mM
4 - 3.96 MHz; 4 ms




**No $C_2$ Swan bands** (or very dim)

**CN emission**
(presence of dissolved air)

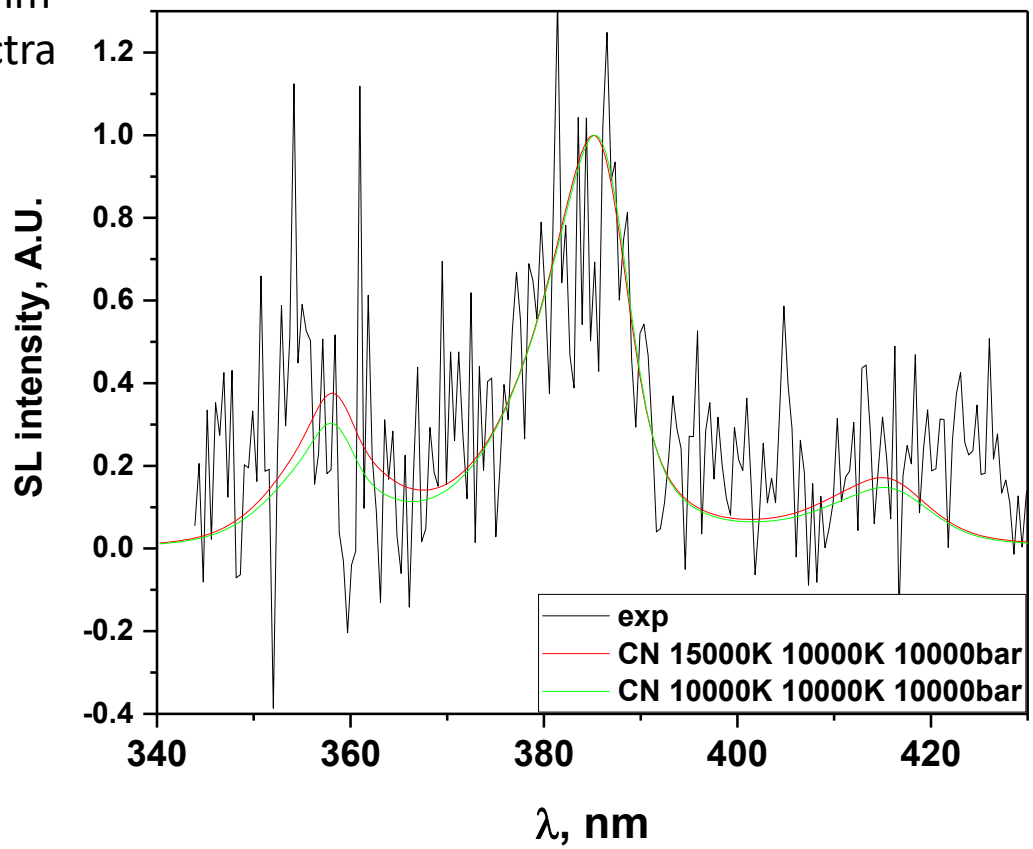
CN emission previously studied at 362 kHz in water saturated with Ar – $CO_2$ – $N_2$ mixtures:

$CO_2 \rightarrow))) \rightarrow CO+O$

$N_2+CO \rightarrow))) \rightarrow CN+NO$

# CN emission

2-propanol 3.7 mM, Ar-20%$O_2$
4 - 3.96 MHz; 4 ms
150blz500, slit 150 µm
spectral resolution 3,3 nm
Average of 6 900-s spectra




Need to further optimize the conditions of observation of CN

# Conclusion

- HIFU SL spectra of aqueous solutions at 4 MHz are very dim but can be measured in vobulated mode
- Different continuum shape: very strong UV part
- Usual molecular emissions even broader than at HF
- CN may be a candidate of choice for fitting but conditions of observation must be optimised

# Thank you
# for your attention!

# Thank you
# for the organisation!

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