

# Performances of a Polaritonic Refrigerant

Maxime Richard<sup>1</sup>

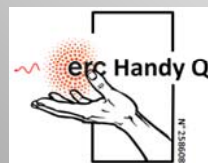
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<sup>3</sup> *Institut für Festkörperphysik, Universität Bremen, Otto-Hahn-Allee, 28359 Bremen, Germany*

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# Outline

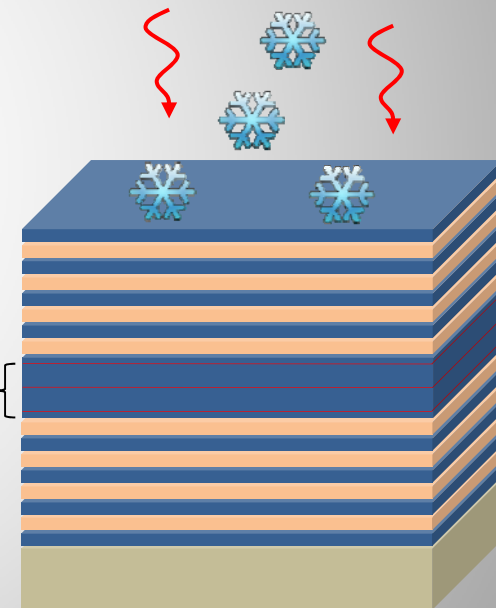


1- Can we use polariton fluids to cool down solids ?

Front mirror

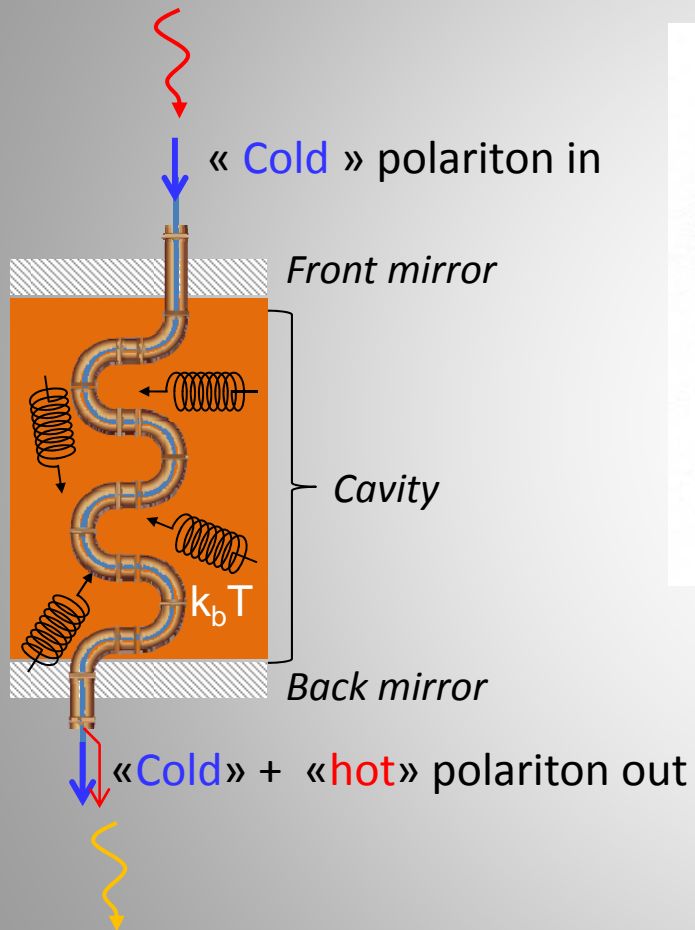
Cavity

Back mirror

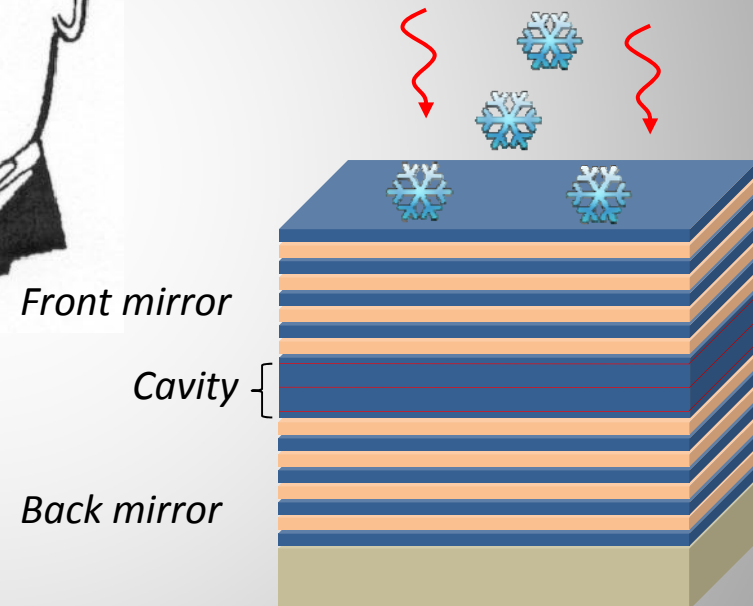


Microcavity in the strong coupling regime

# Outline



1- Can we use polariton fluids to cool down solids ?



Microcavity in the strong coupling regime

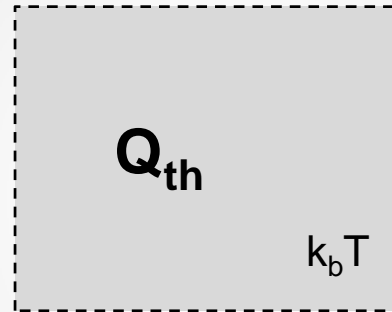
2- What kind of heat exchange fluid is that ?

# Outline

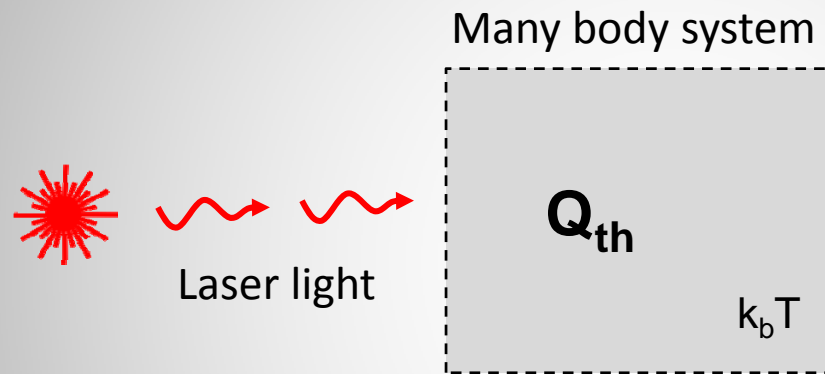
**0 – Back to the principle of cooling many body systems with light**

# Principle of optical cooling

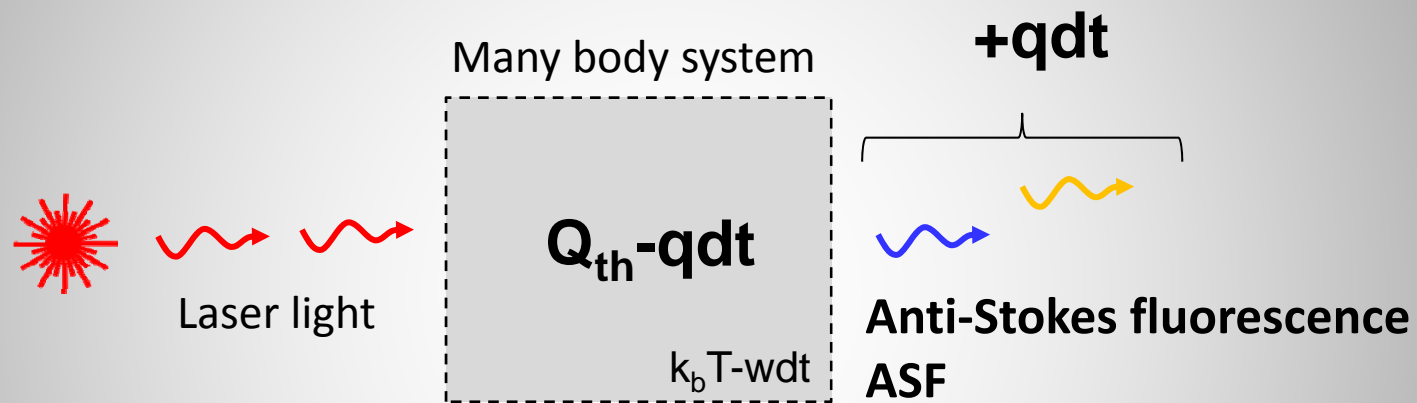
Many body system



# Principle of optical cooling



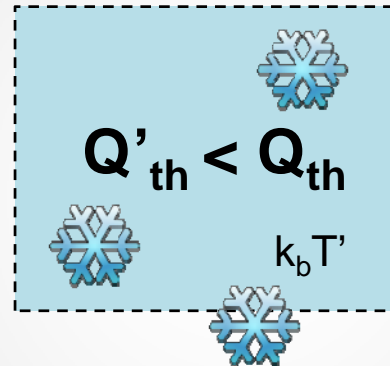
# Principle of optical cooling



# Principle of optical cooling



Many body system

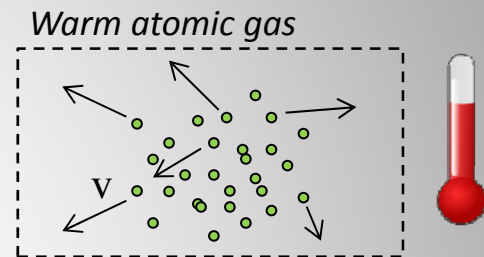




# Principle of optical cooling

## Doppler cooling

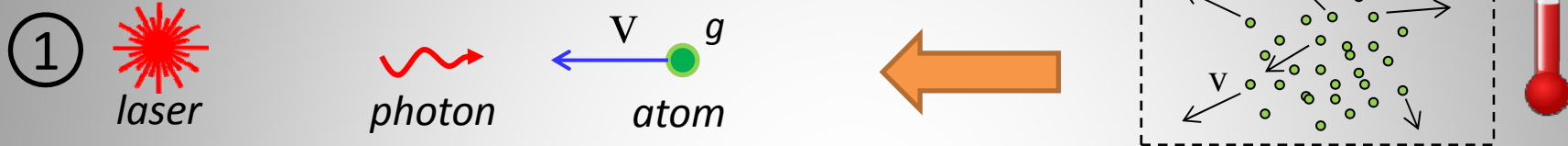
Cooling the translational degrees of freedom



# Principle of optical cooling

## Doppler cooling

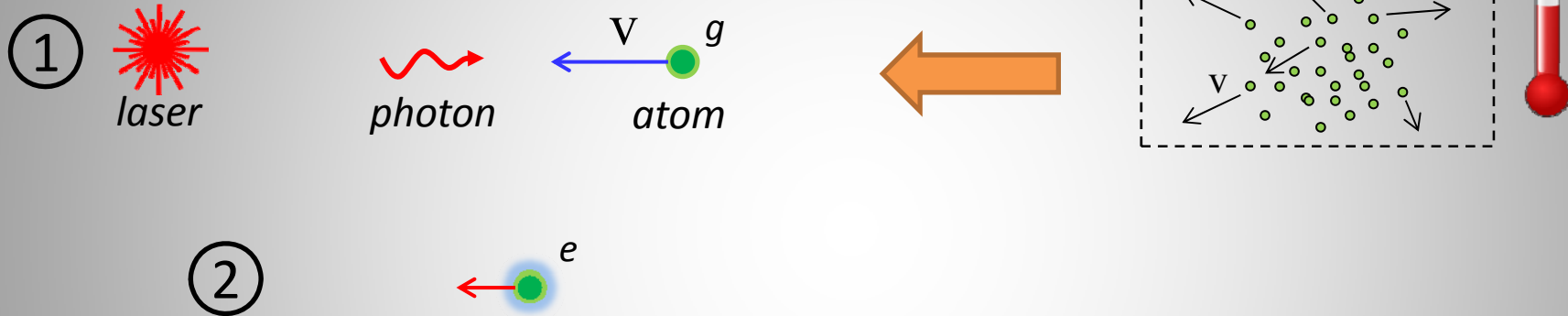
Cooling the translational degrees of freedom



# Principle of optical cooling

## Doppler cooling

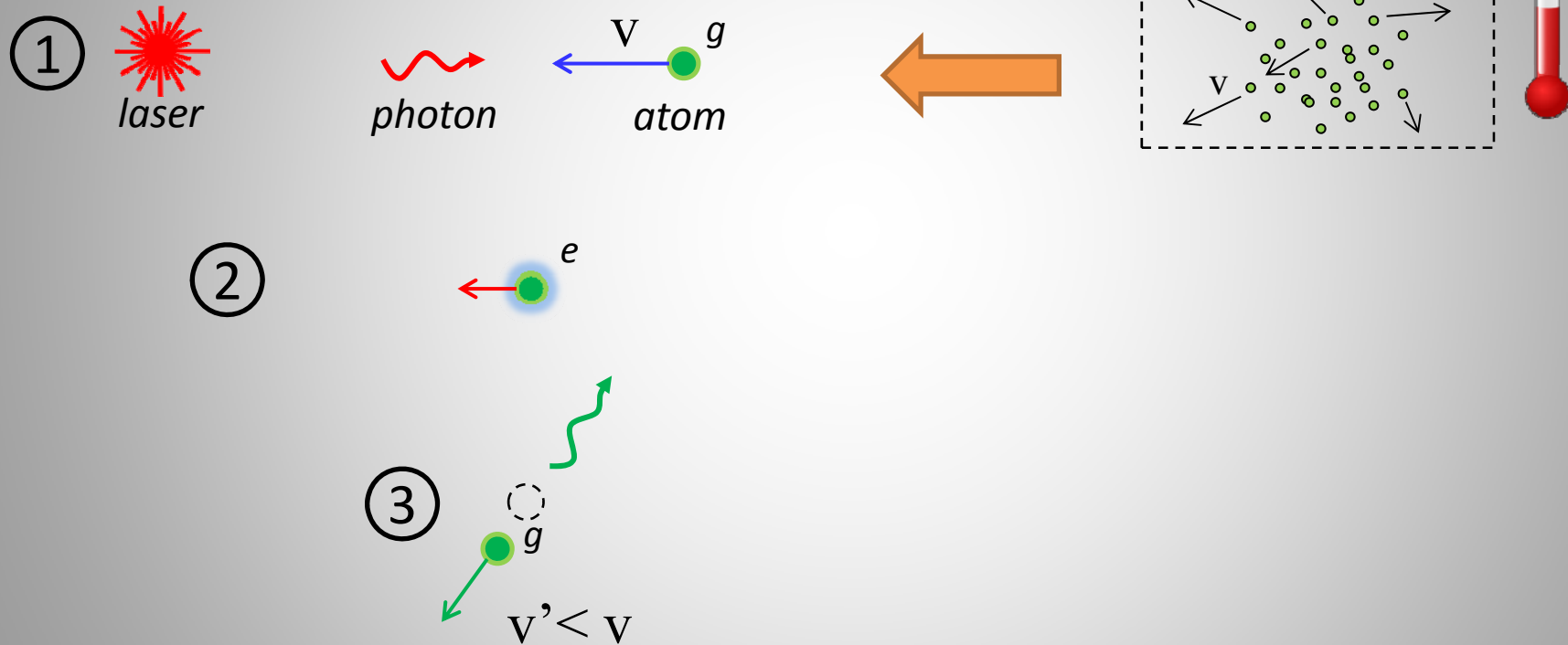
Cooling the translational degrees of freedom



# Principle of optical cooling

## Doppler cooling

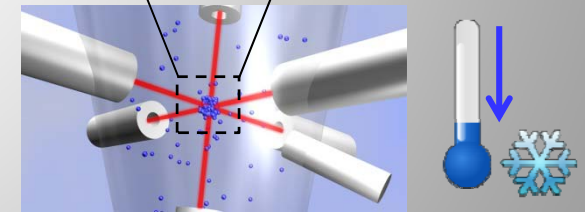
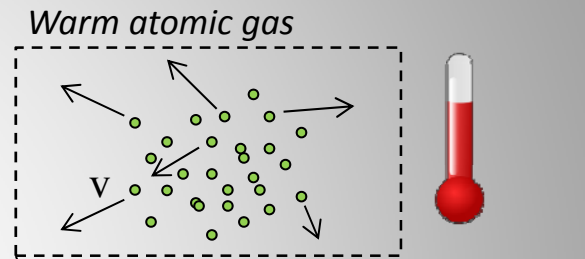
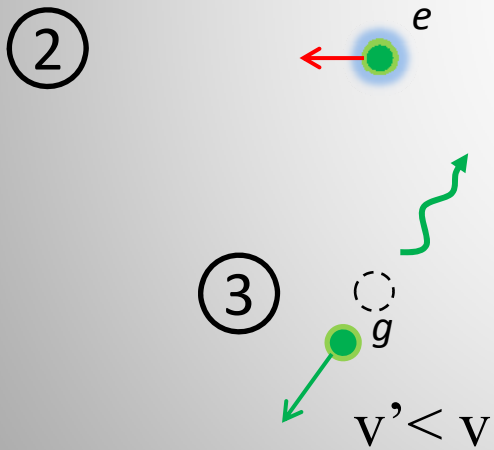
Cooling the translational degrees of freedom



# Principle of optical cooling

## Doppler cooling

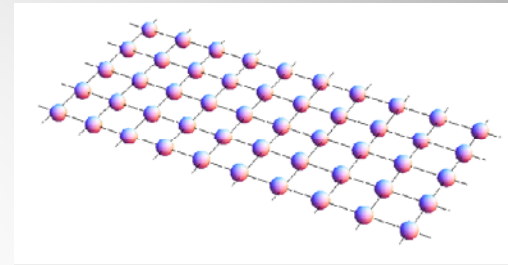
Cooling the translational degrees of freedom



*Schematic arrangement of a doppler cooling setup*

# Principle of optical cooling

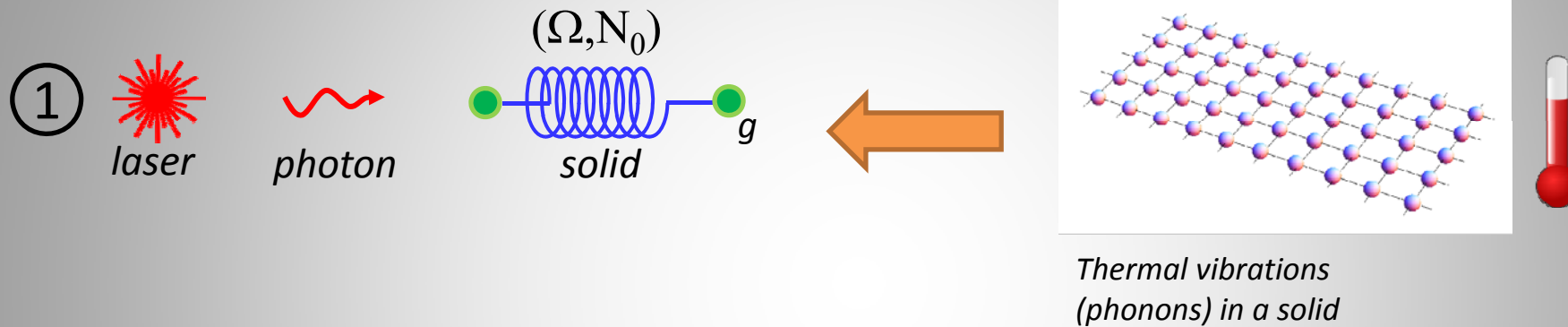
**Cooling by anti-Stokes fluorescence (ASF)**  
Cooling the vibrational degrees of freedom



*Thermal vibrations  
(phonons) in a solid*

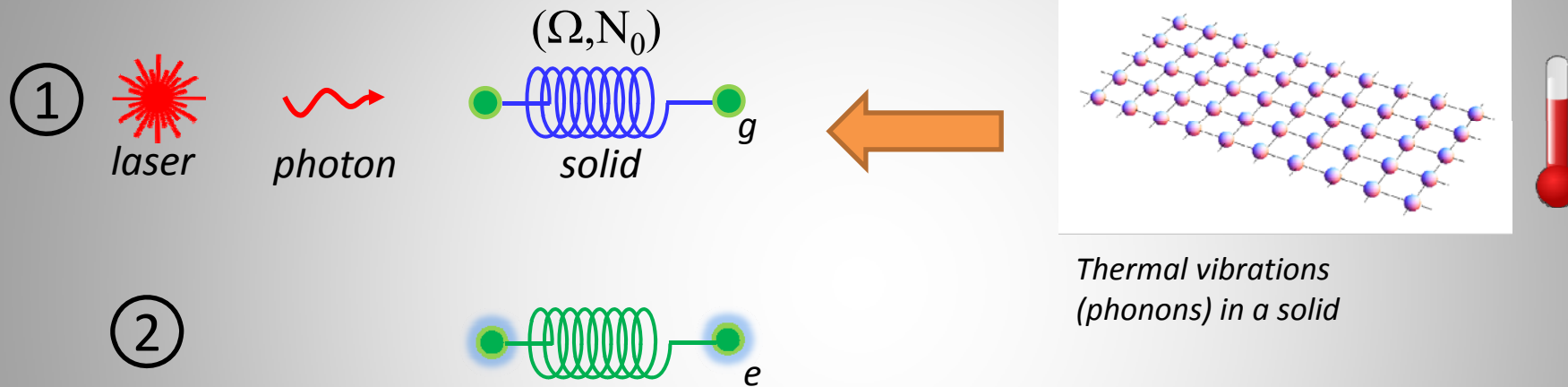
# Principle of optical cooling

**Cooling by anti-Stokes fluorescence (ASF)**  
Cooling the vibrational degrees of freedom



# Principle of optical cooling

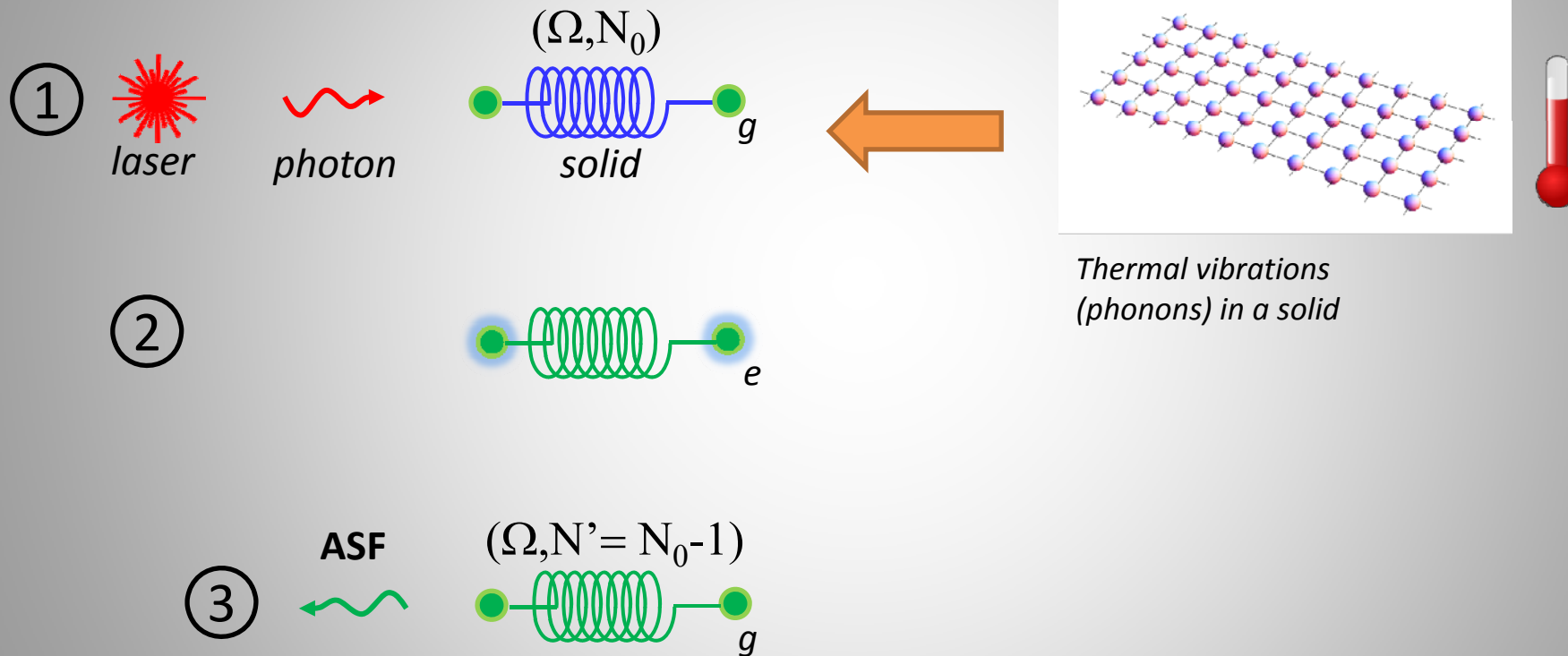
## Cooling by anti-Stokes fluorescence (ASF) Cooling the vibrational degrees of freedom





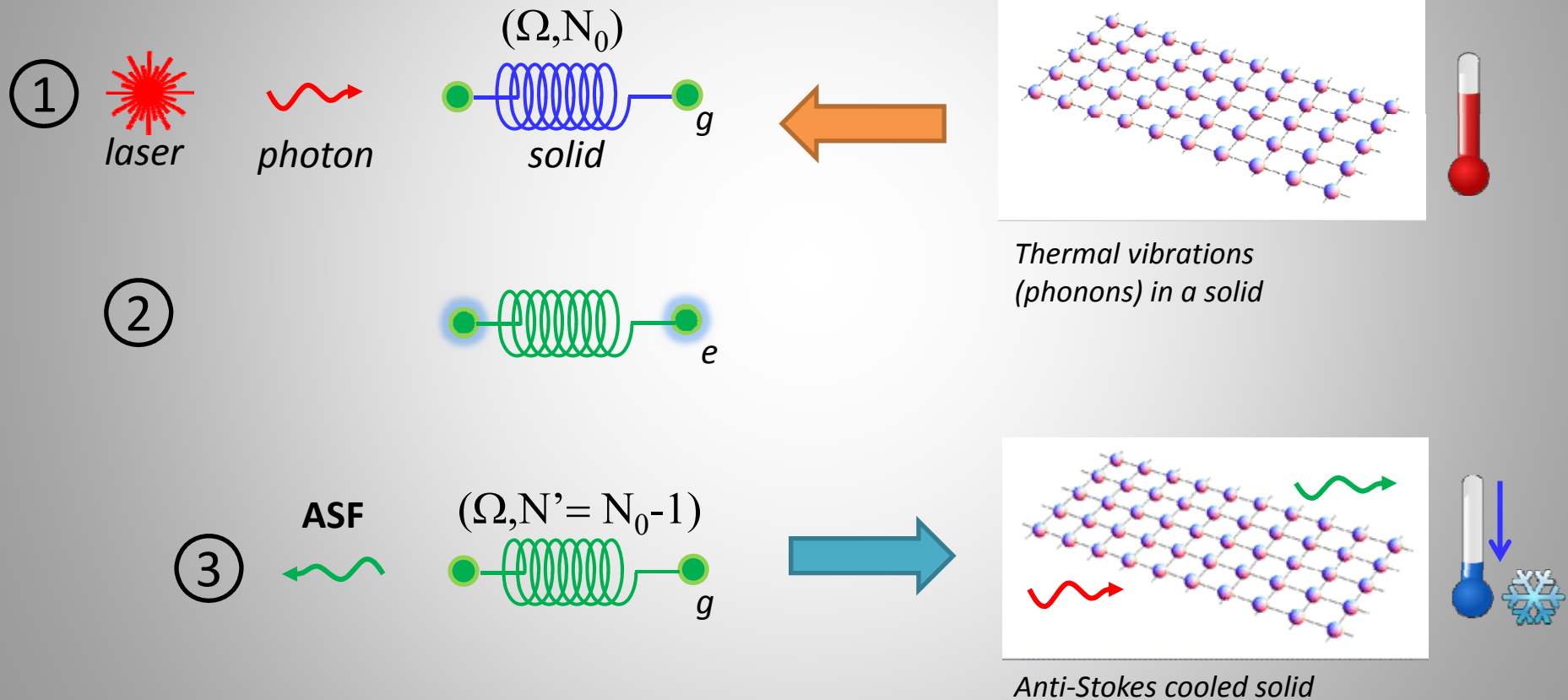
# Principle of optical cooling

## Cooling by anti-Stokes fluorescence (ASF) Cooling the vibrational degrees of freedom



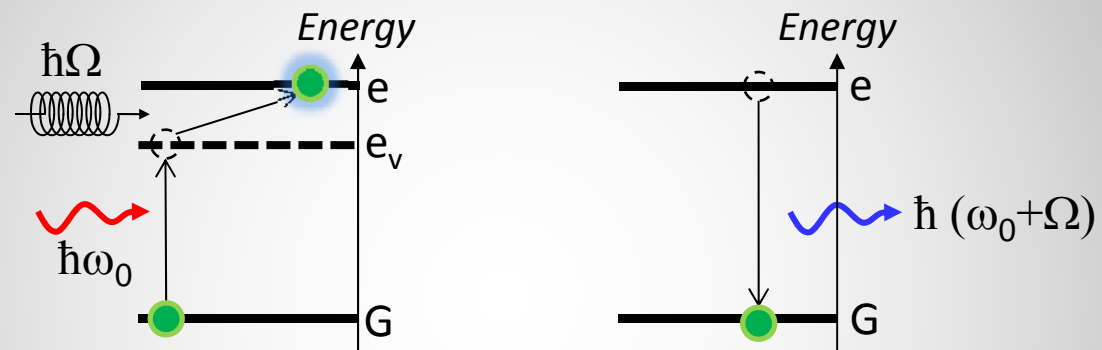
# Principle of optical cooling

## Cooling by anti-Stokes fluorescence (ASF) Cooling the vibrational degrees of freedom

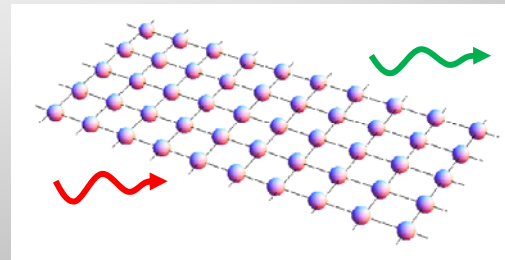


# Principle of optical cooling

## 1- Anti-Stokes fluorescence mechanism

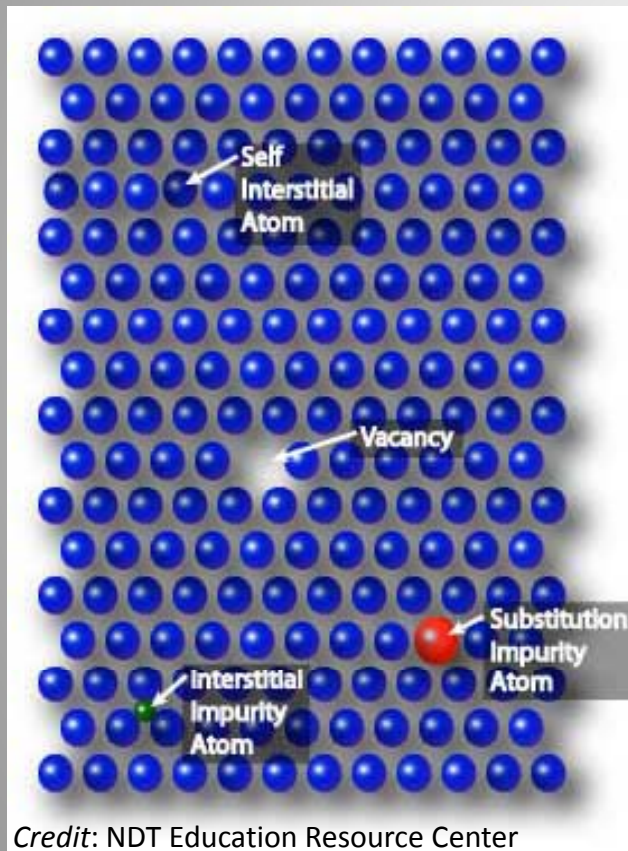


Removes  $\hbar\Omega$  per scattering event from the thermal phonons bath



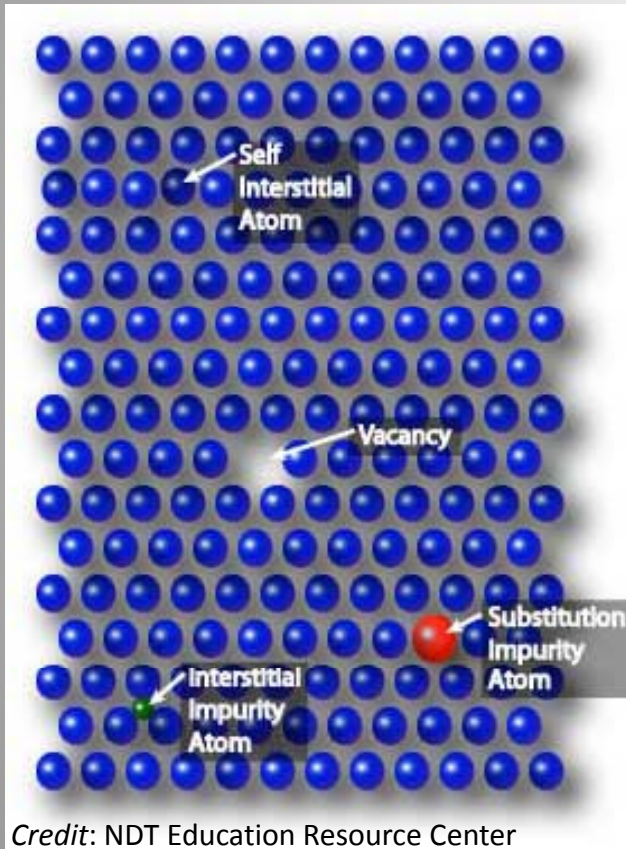
# Principle of optical cooling

Realistic solids have defects

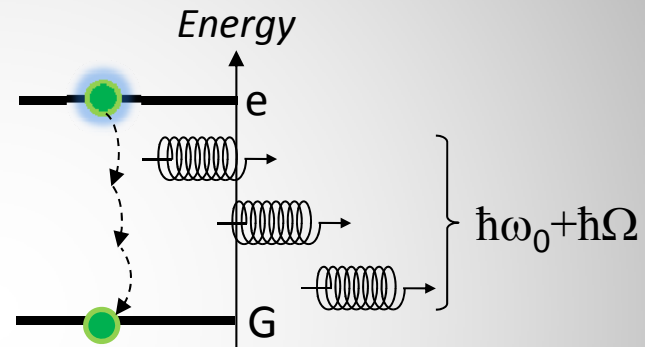


# Principle of optical cooling

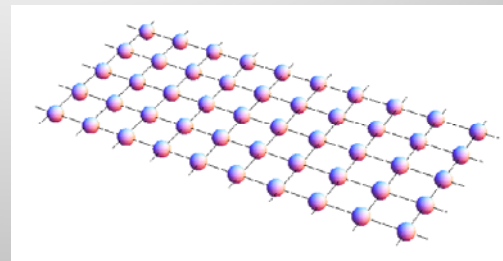
Realistic solids have defects



## 2- Non-radiative recombination



Adds  $\hbar\omega_0$  per scattering event to the thermal phonons bath



Vibrations (phonons) in a lattice

## Requirements to achieve net cooling power in solids

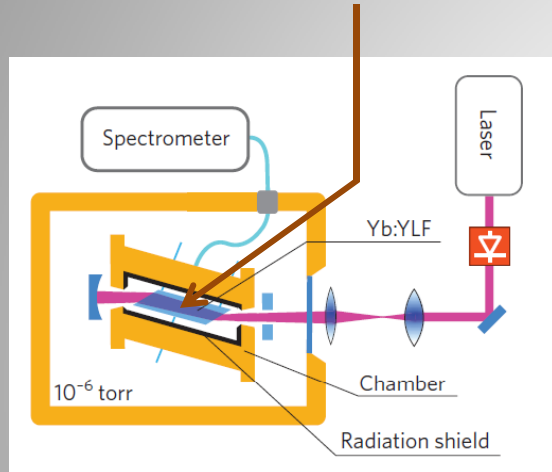
To achieve  power in solids you need :

→ Largest possible quantum efficiency  $\eta$

→ Largest possible oscillator strength  $f$   
i.e. short radiative lifetime  $\tau$  of e

# atoms embedded in solid matrix

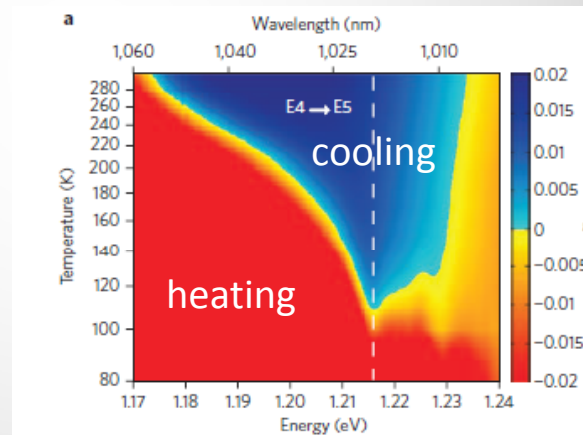
## Ytterbium doped glass



Setup for optical cooling of a  $\text{LiYF}_4:\text{Yb}$  rod [1]

From room temperature  
down to  $T \sim 110\text{K}$  [2]

- Excellent  $\eta$
- Poor  $f$
- Coupling with phonons is of 2<sup>nd</sup> order



Cooling efficiency  
in  $\text{LiYF}_4:\text{Yb}$  under  
optical cooling [1]

[1] D. V. Seletskiy *et al.* Nature Photonics **4** 161 (2010)

[2] D. V. Seletskiy *et al.* Optics Express **19**, 18229 (2011)



# Semiconductor hetero/nanostructures

CdS nanobelts

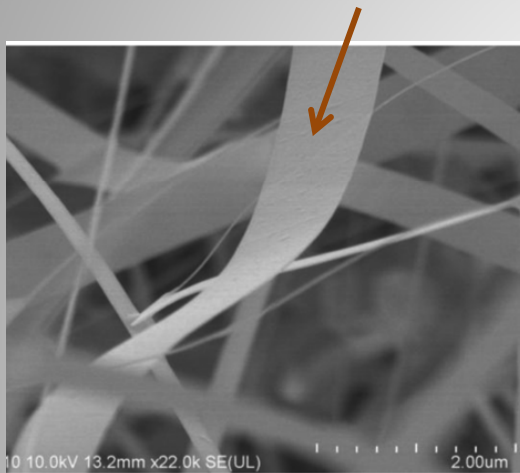
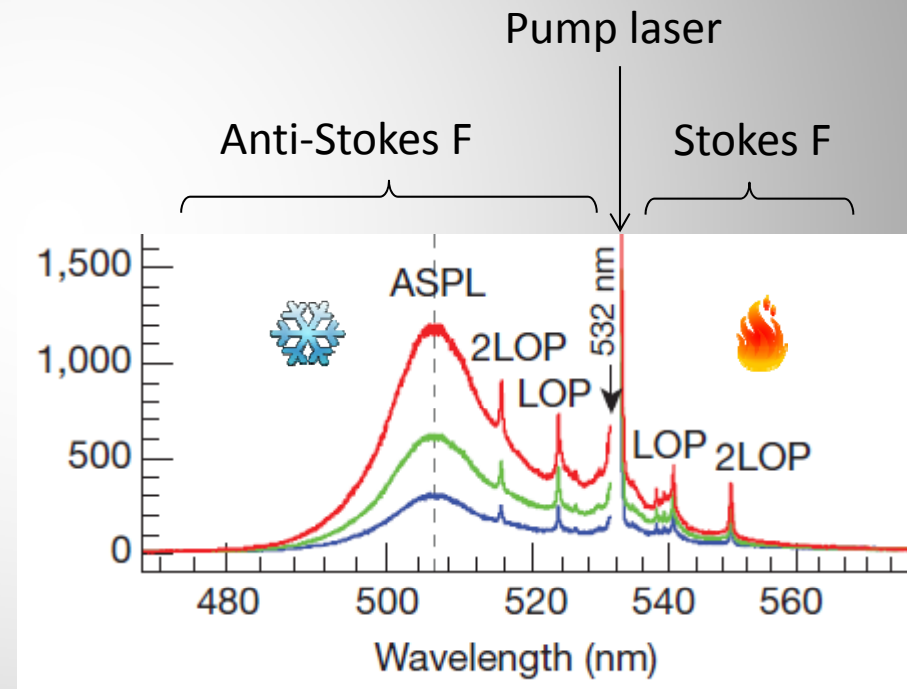


image credit: L. Li et al. Sensors **14**, 7332 (2014)

From room temperature  
down to  $T \sim 260\text{K}$  [3]

- Lower  $\eta$
- **larger  $f$**  (excitonic enhancement [4])
- Coupling with phonons is of 2<sup>nd</sup> order



Raman spectra of a single CdS nanobelt: from ref [3]

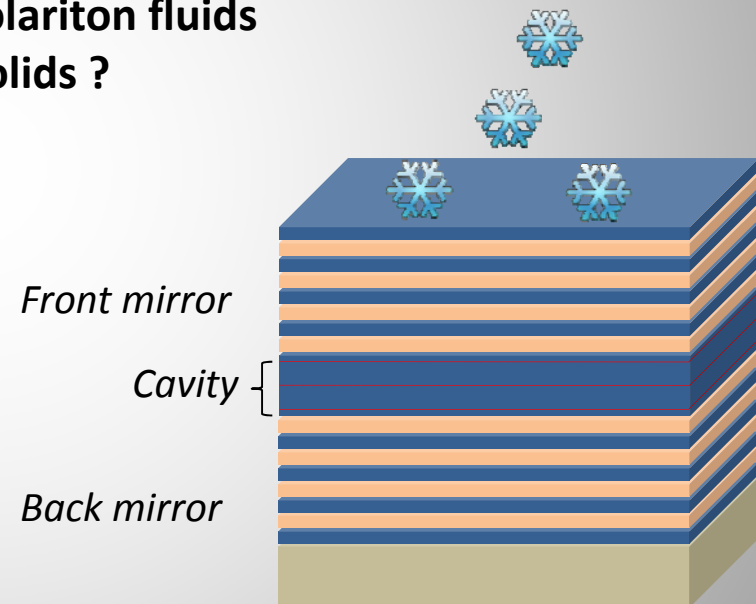
[3] J. Zhang et al. , Nature **493**, 504-508 (2013)

[4] G. Rupper et al. Phys. Rev. Lett. **97** 117401 (2006)



# Outline

**1- Can we use polariton fluids to cool down solids ?**

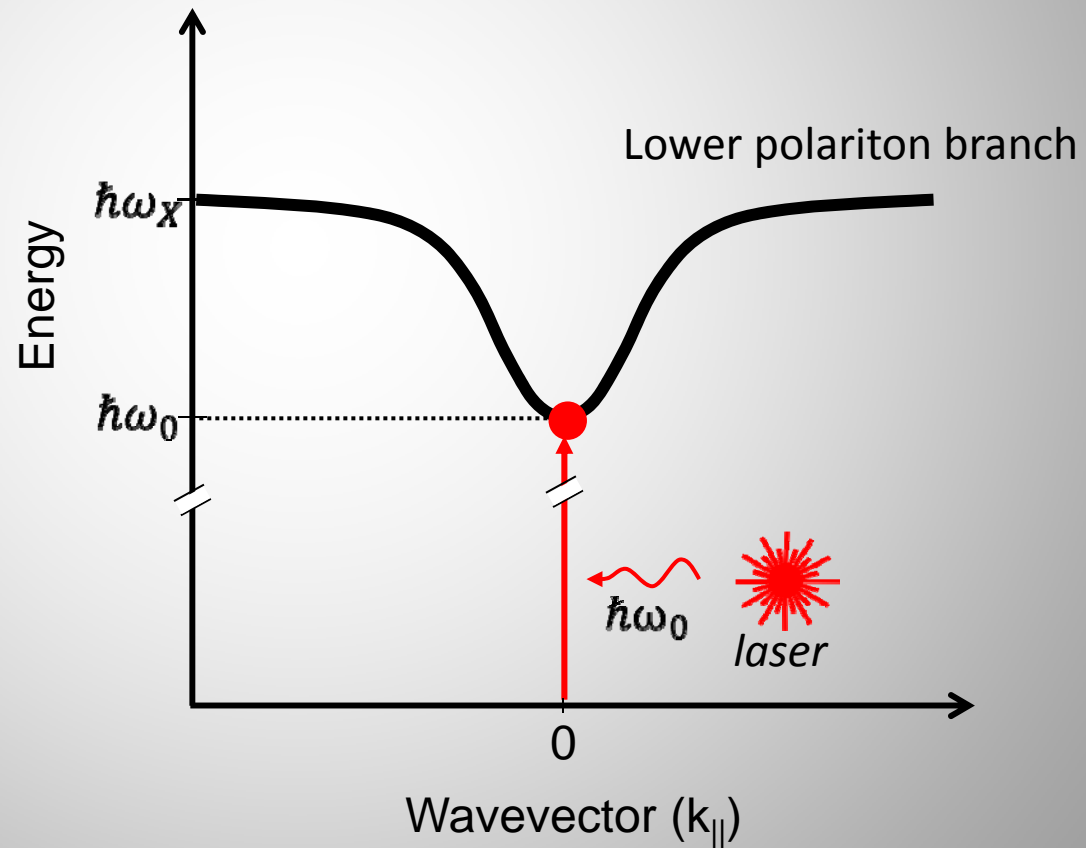


Microcavity in the strong coupling regime

# Cooling a solid-state $\mu$ -cavity with polaritons

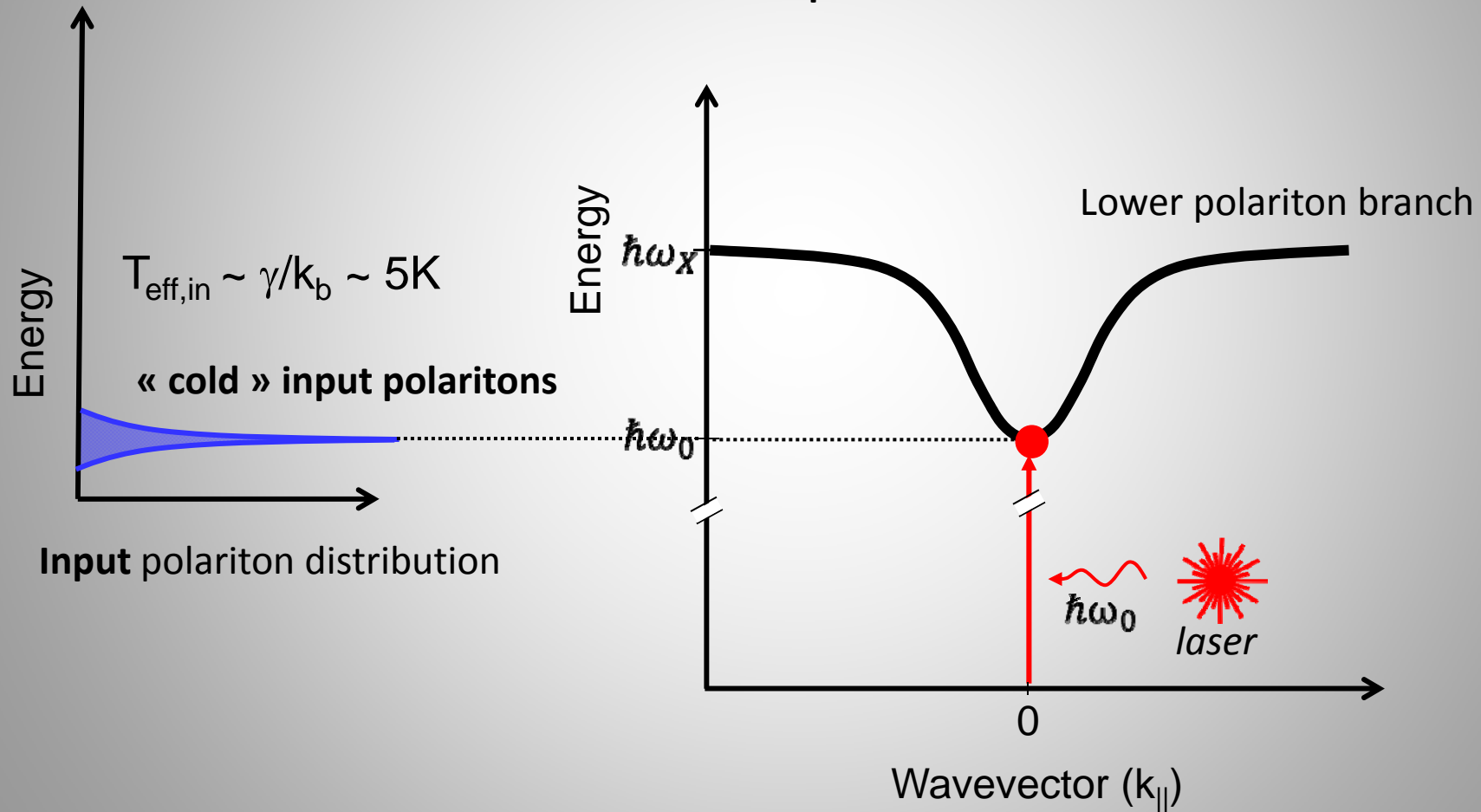
Polariton anti-Stokes fluorescence

**Principle :**



# Cooling a solid-state $\mu$ -cavity with polaritons

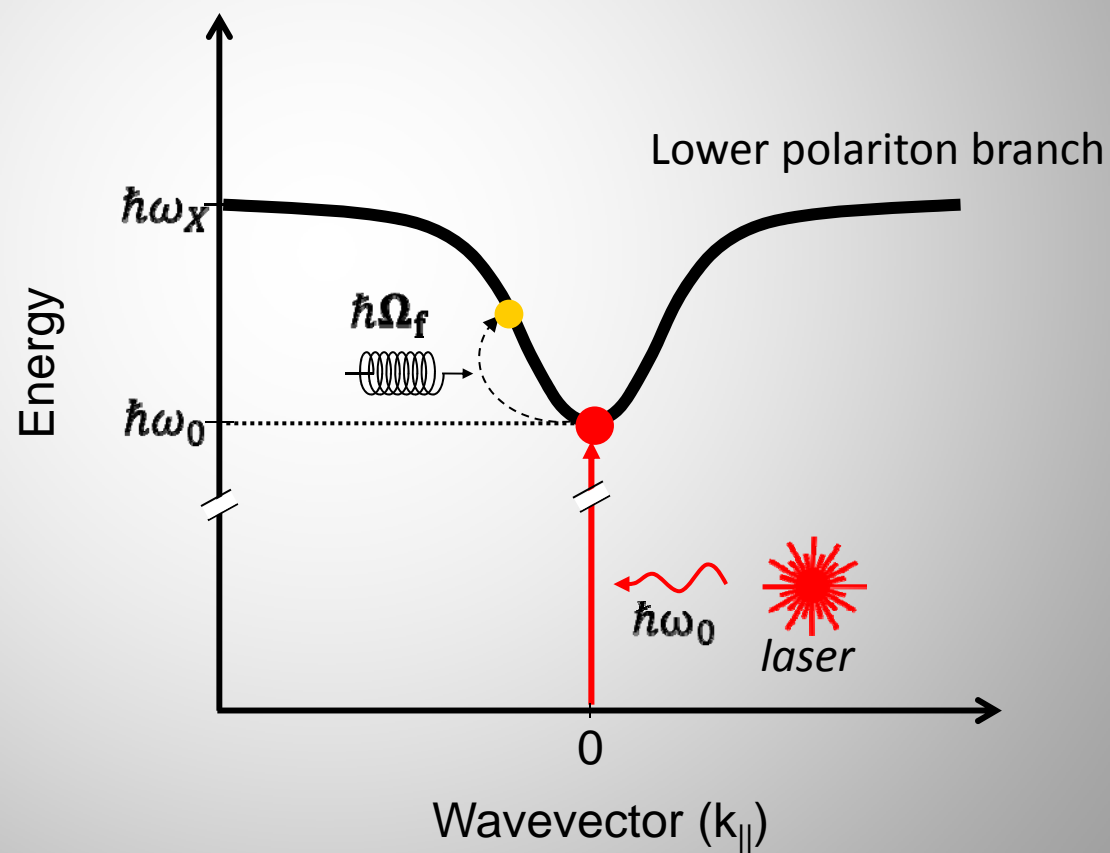
Polariton anti-Stokes fluorescence  
**Principle :**



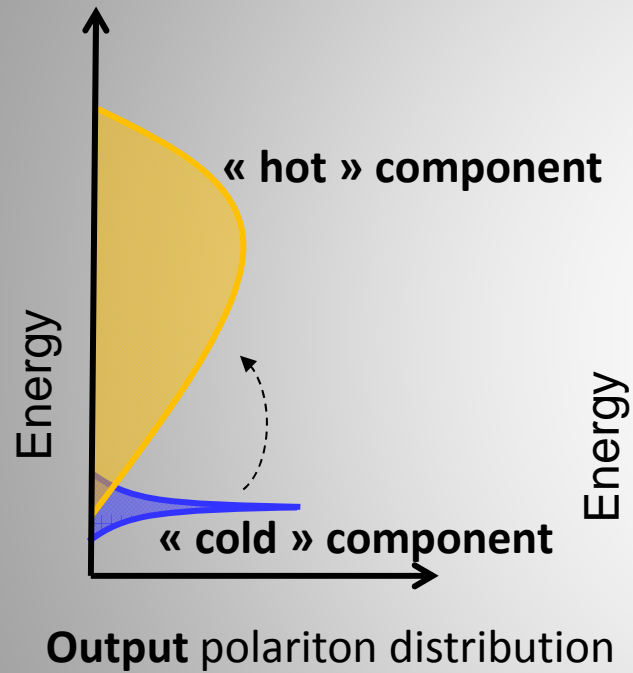
# Cooling a solid-state $\mu$ -cavity with polaritons

Polariton anti-Stokes fluorescence

**Principle :**



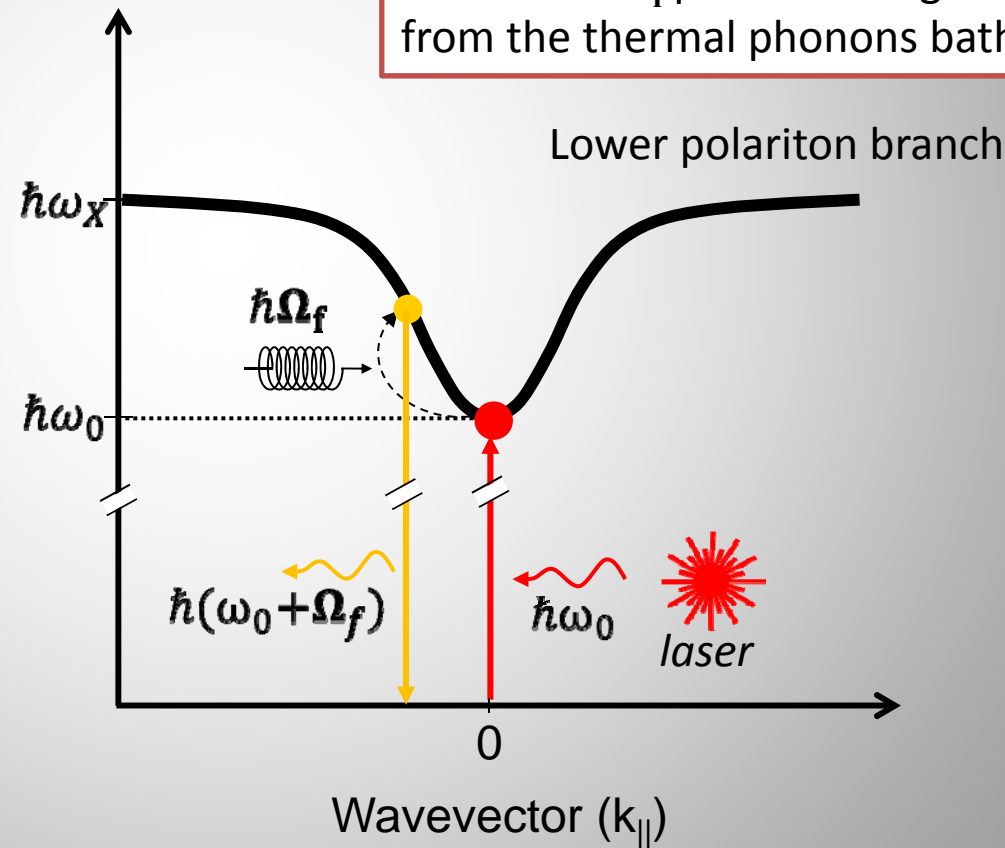
# Cooling a solid-state $\mu$ -cavity with polaritons



Polariton anti-Stokes fluorescence

Principle :

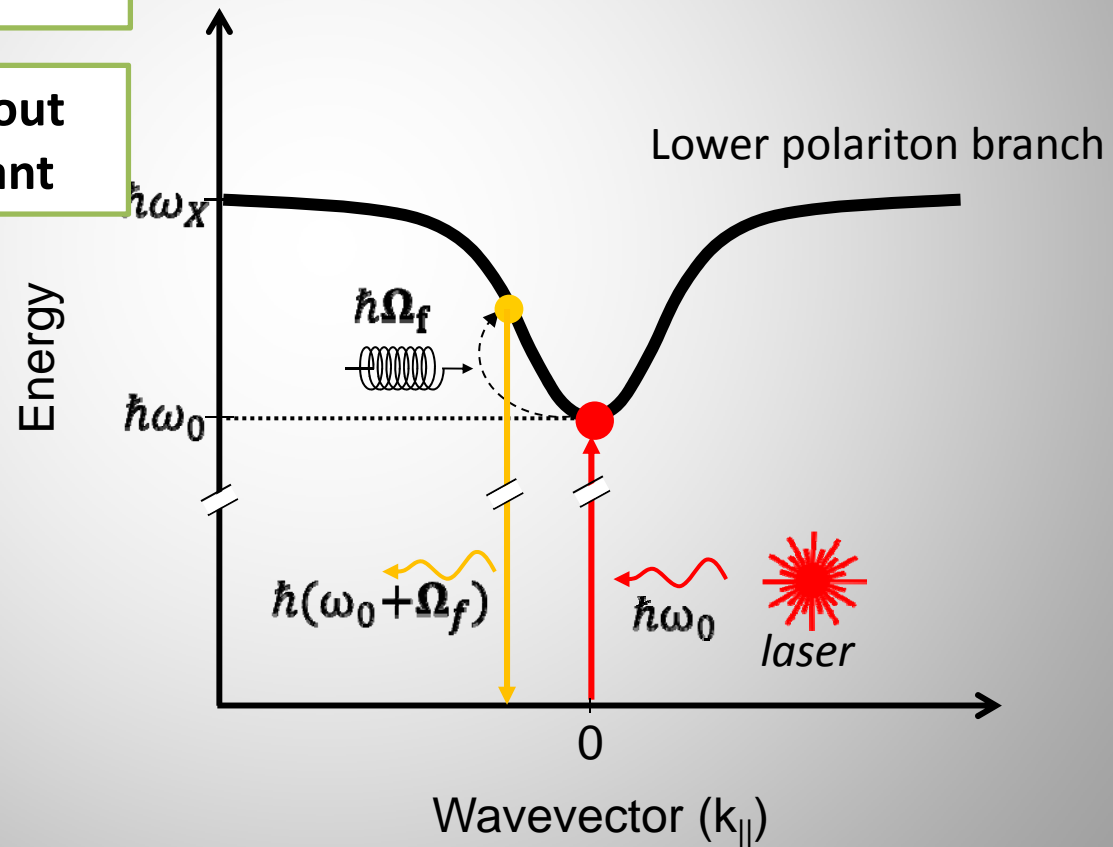
Removes  $\hbar\Omega_f$  per scattering event from the thermal phonons bath



# Cooling a solid-state $\mu$ -cavity with polaritons

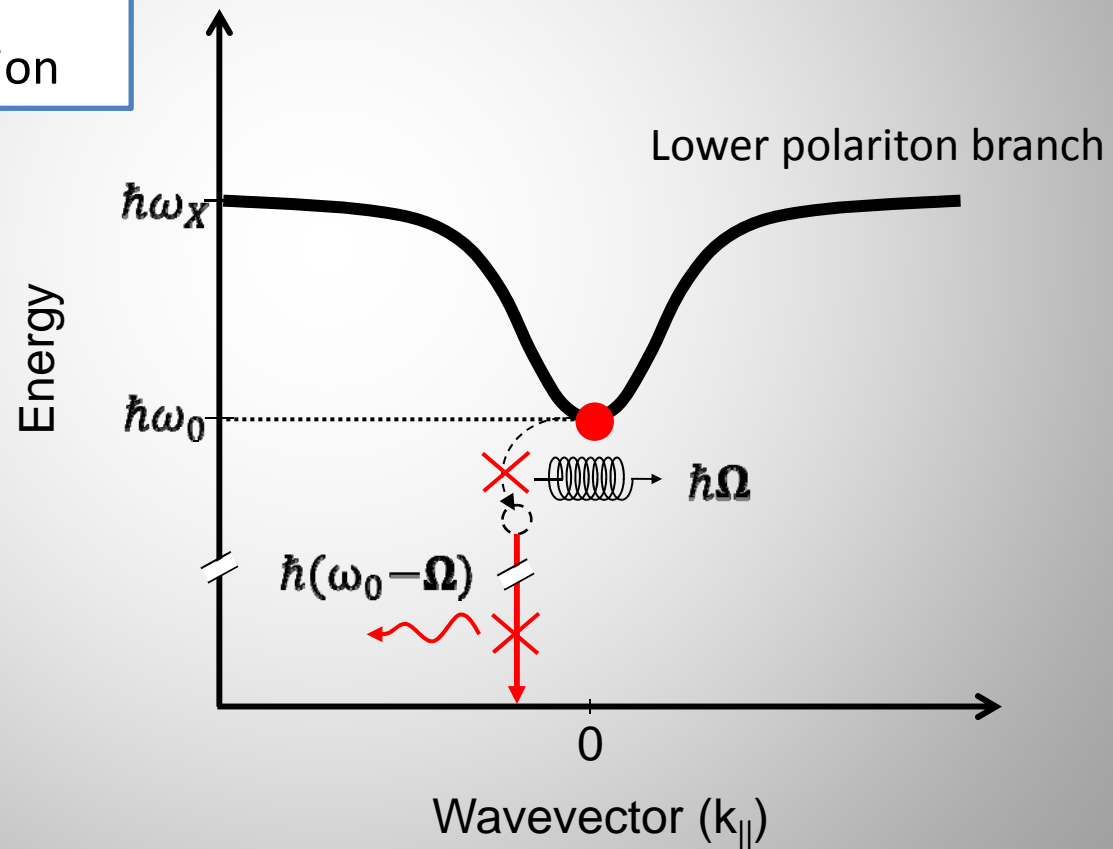
- **Excitonic fraction**  
→ Large, 1<sup>st</sup> order coupling with phonons [5]

→ Expt. Realization of an out of equilibrium refrigerant



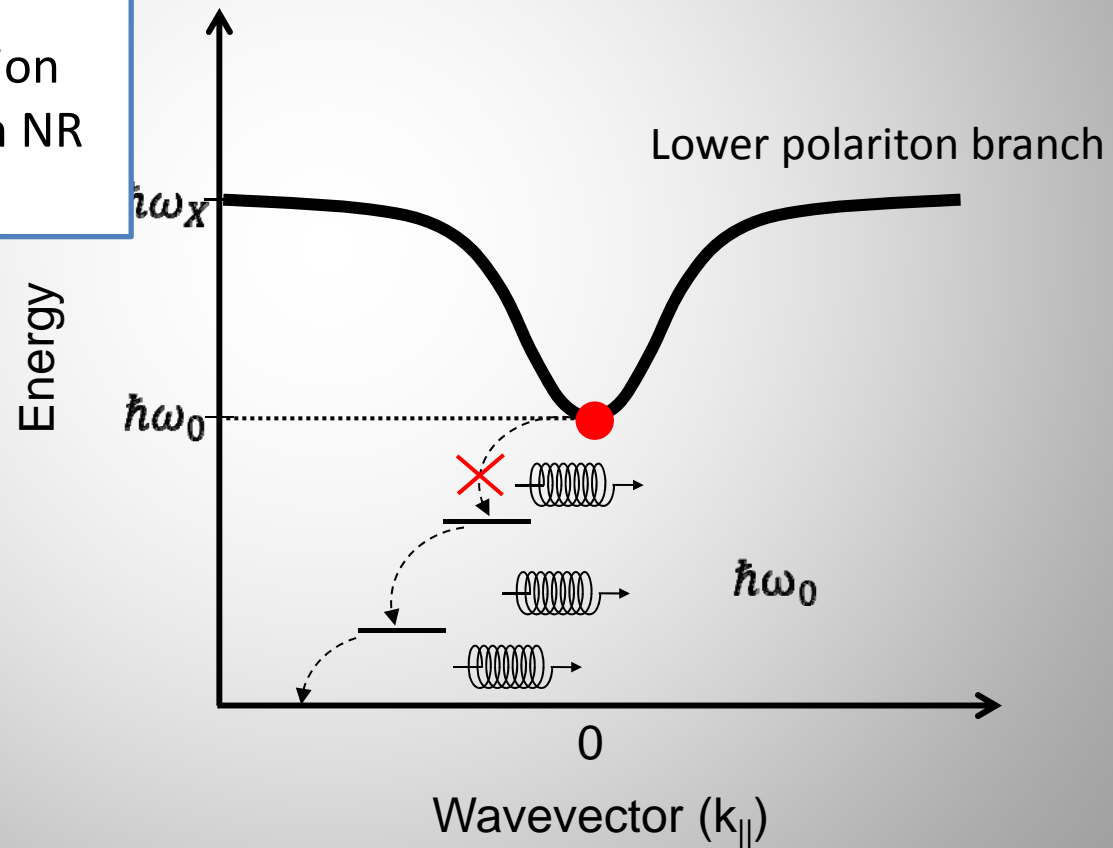
# Cooling a solid-state $\mu$ -cavity with polaritons

- **Polaritonic dispersion**  
→ quenched Stokes emission



# Cooling a solid-state $\mu$ -cavity with polaritons

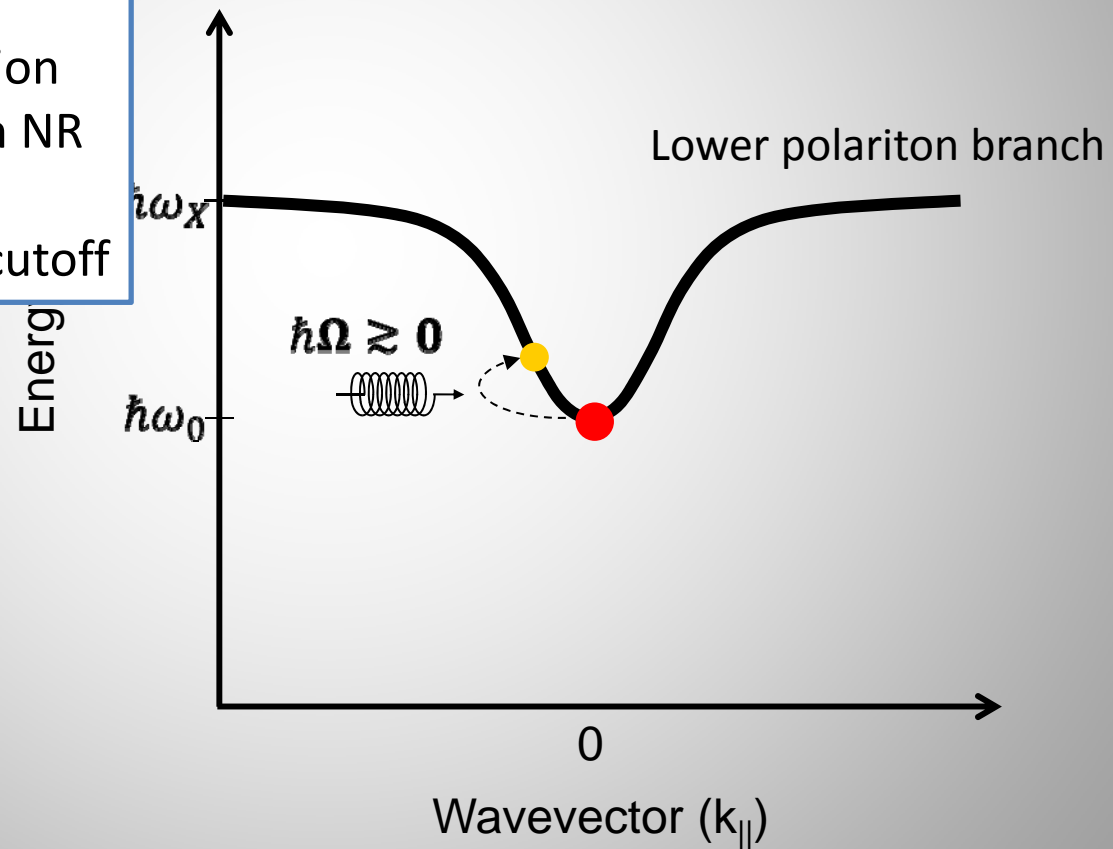
- **Polaritonic dispersion**
  - quenched Stokes emission
  - quenched coupling with NR point defects





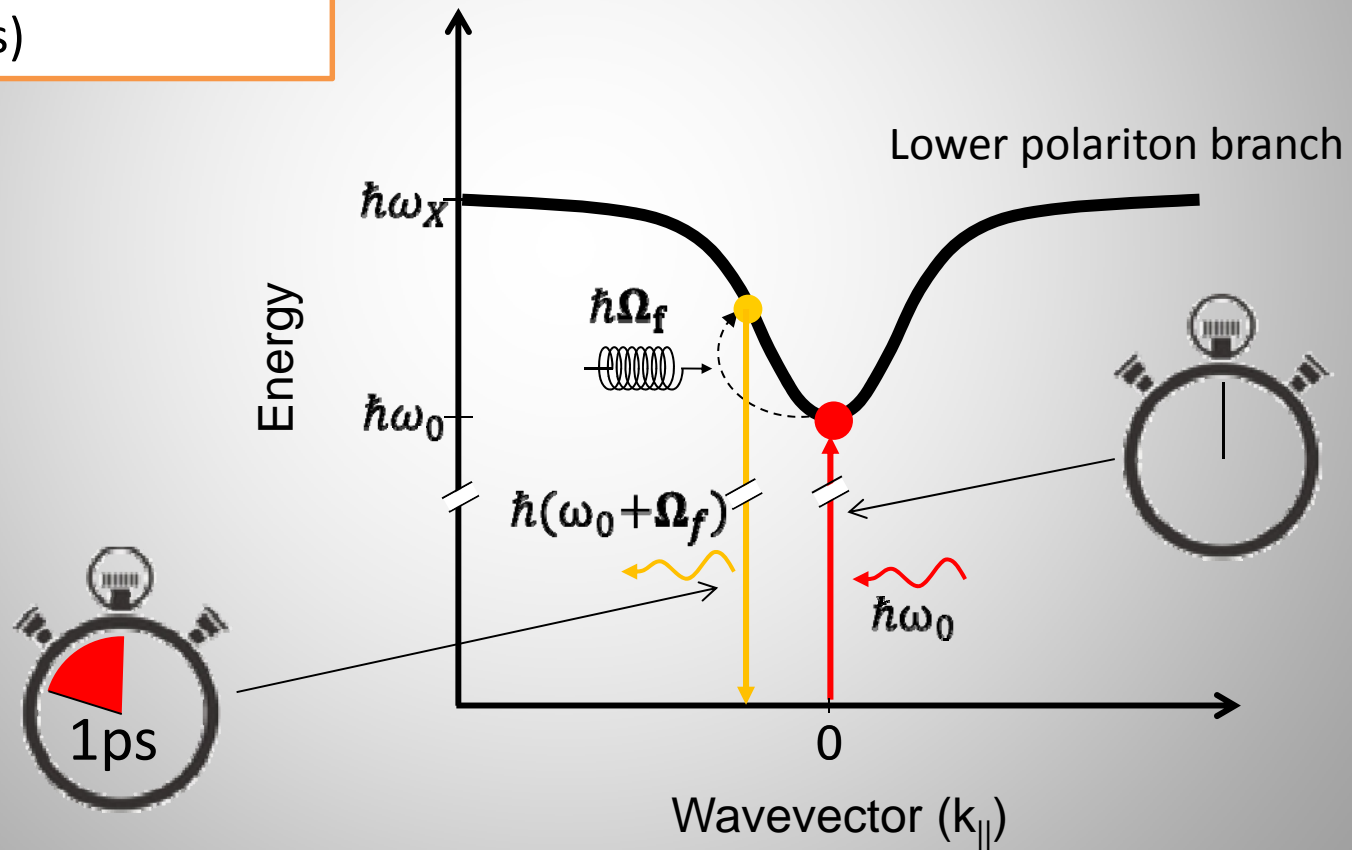
## Cooling a solid-state $\mu$ -cavity with polaritons

- **Polaritonic dispersion**
  - quenched Stokes emission
  - quenched coupling with NR point defects
  - ~ No low temperature cutoff



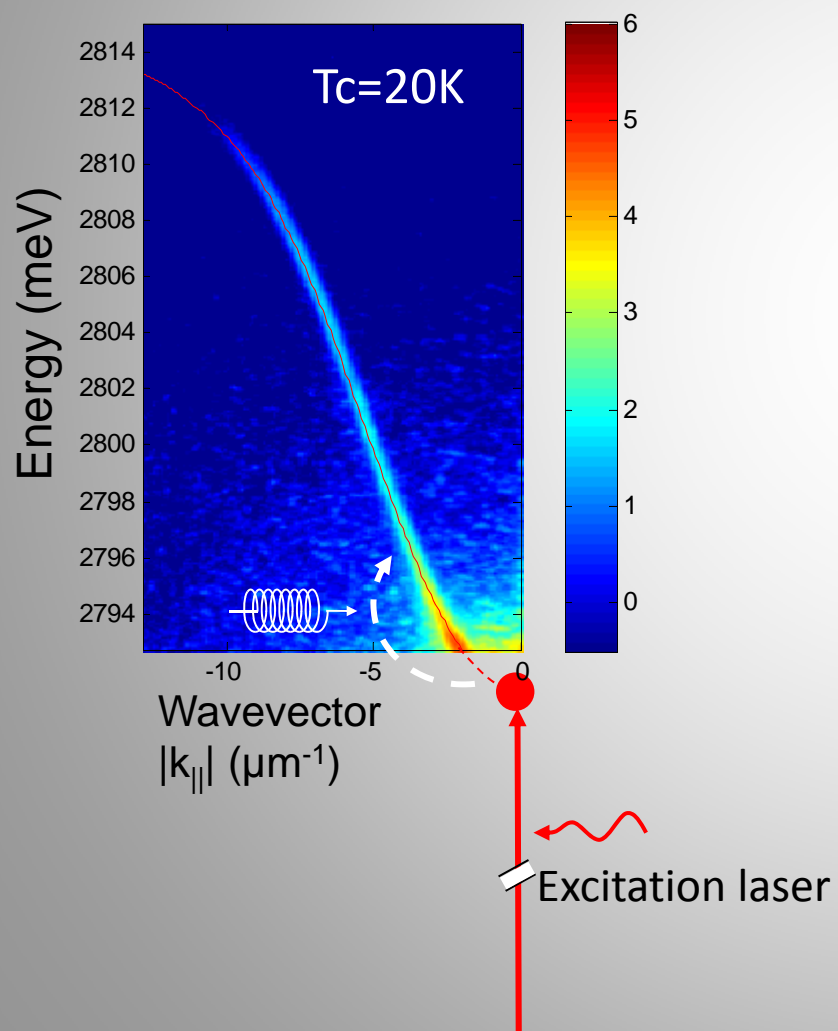
# Cooling a solid-state $\mu$ -cavity with polaritons

- **Photonic fraction**  
→ **Extra short** radiative lifetime, i.e. ultrafast cooling dynamics ( $\sim 1\text{ps}$ )



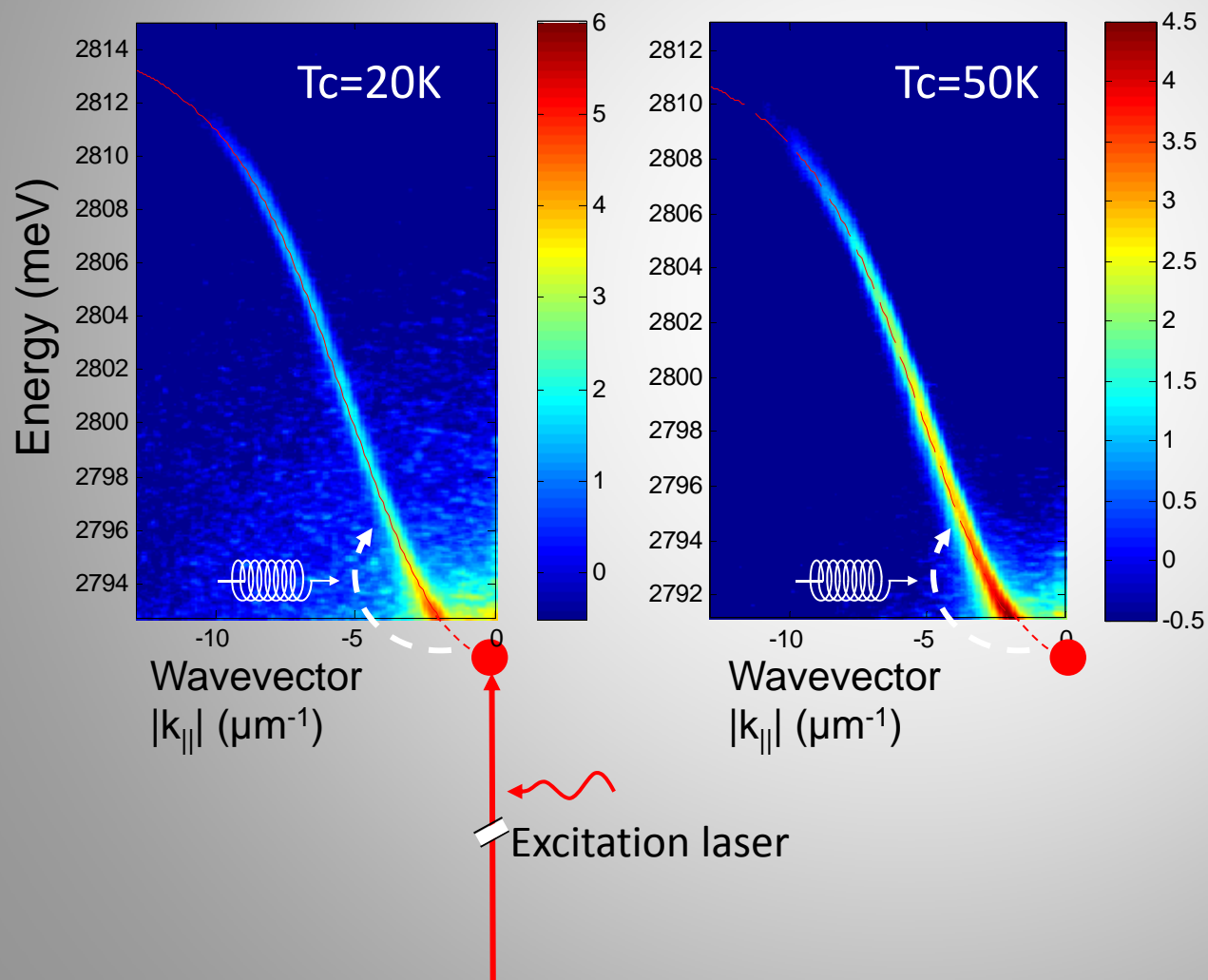
# anti-Stokes fluorescence of polaritons

Measured ASF intensity cts/s (log scale)



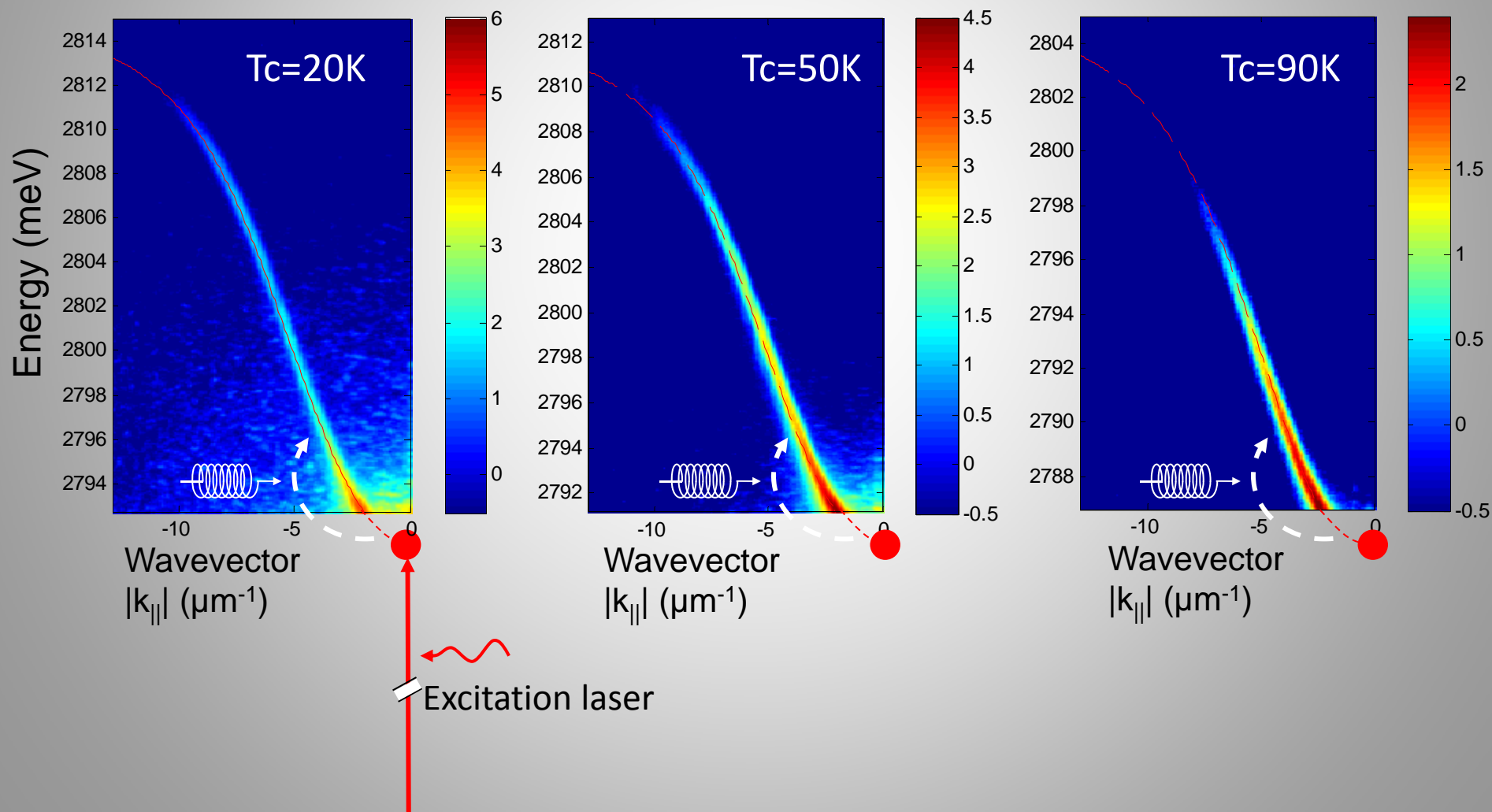
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Measured ASF intensity cts/s (log scale)



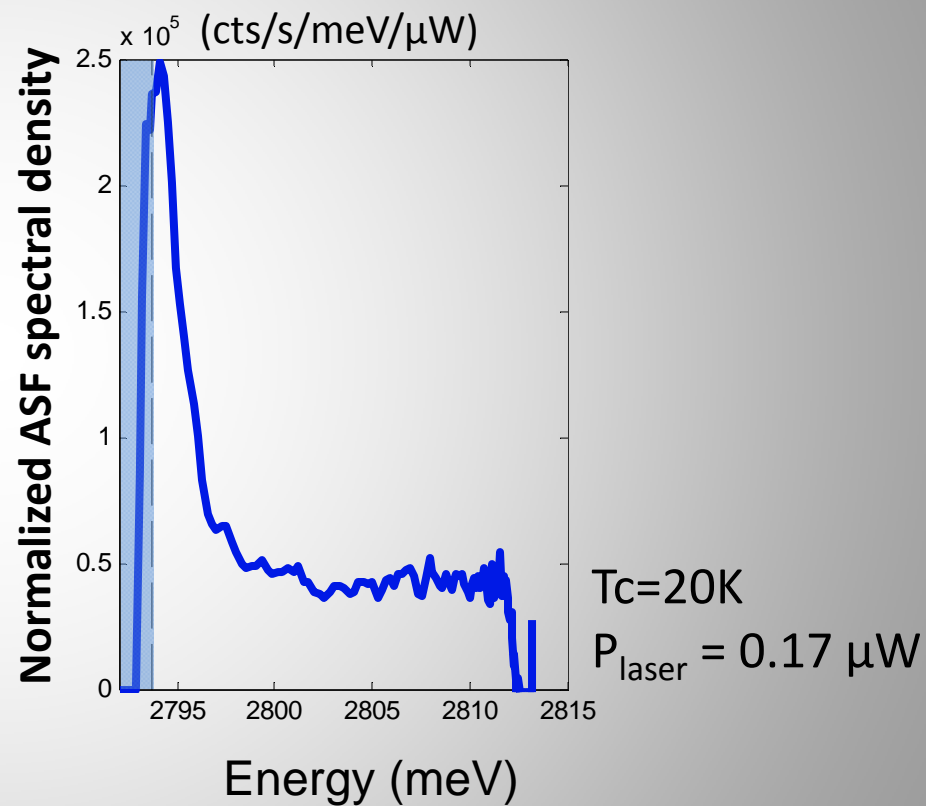
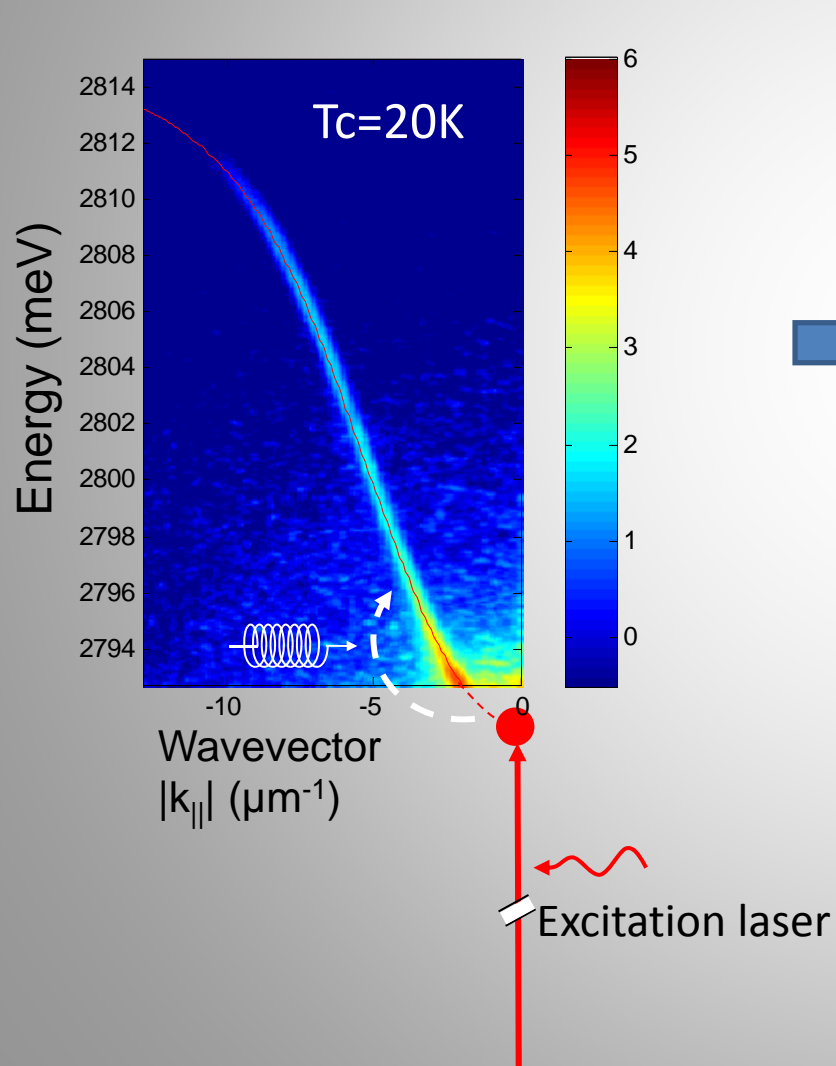
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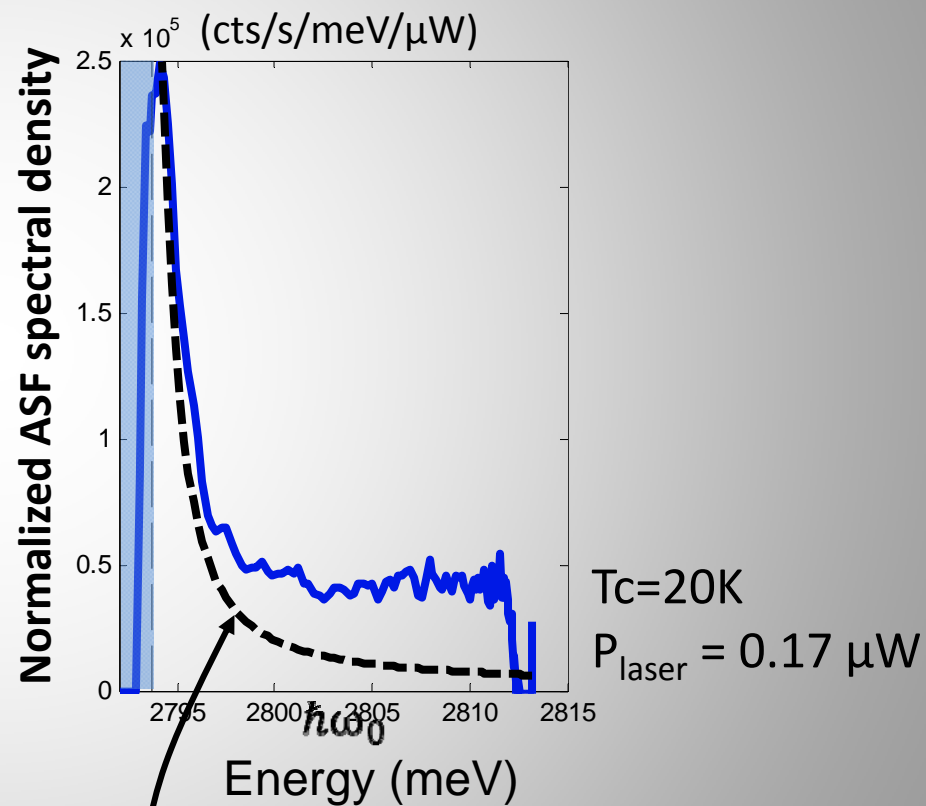
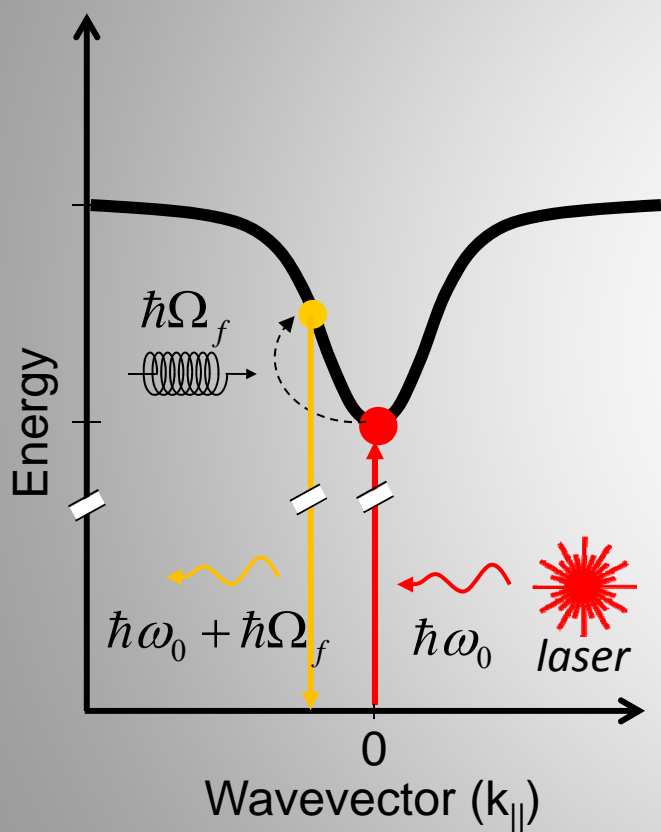
# anti-Stokes fluorescence of polaritons

ASF intensity cts/s (log scale)



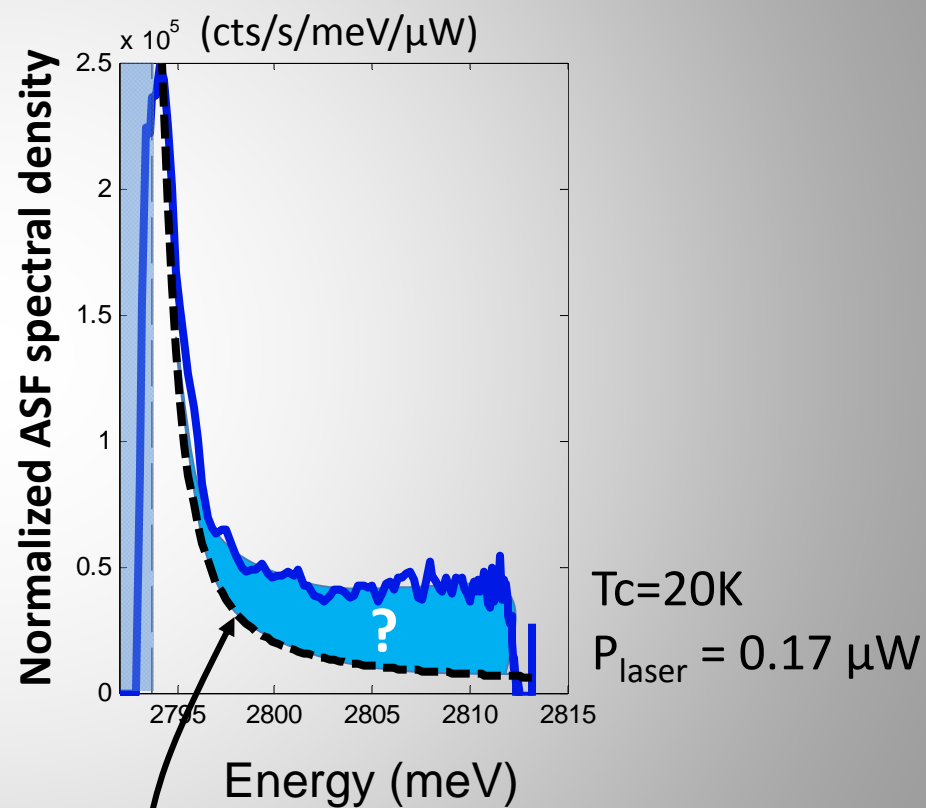
# anti-Stokes fluorescence of polaritons

Fast cooling mechanism ❄️



Theoretical calculation of the fast ASF spectrum (FG rule)

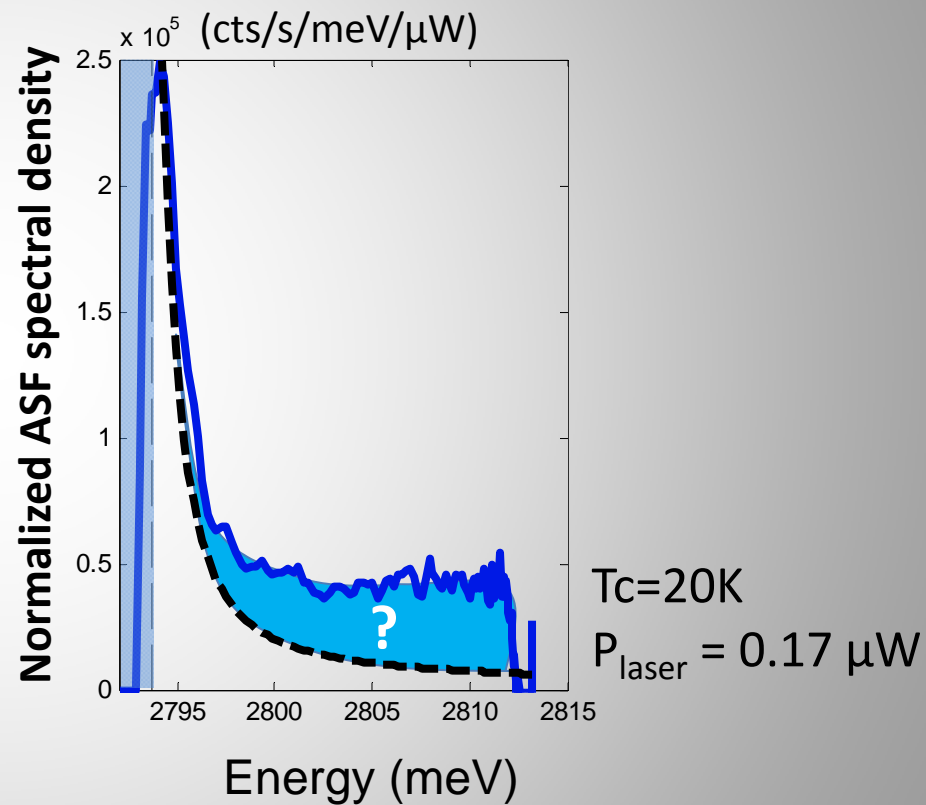
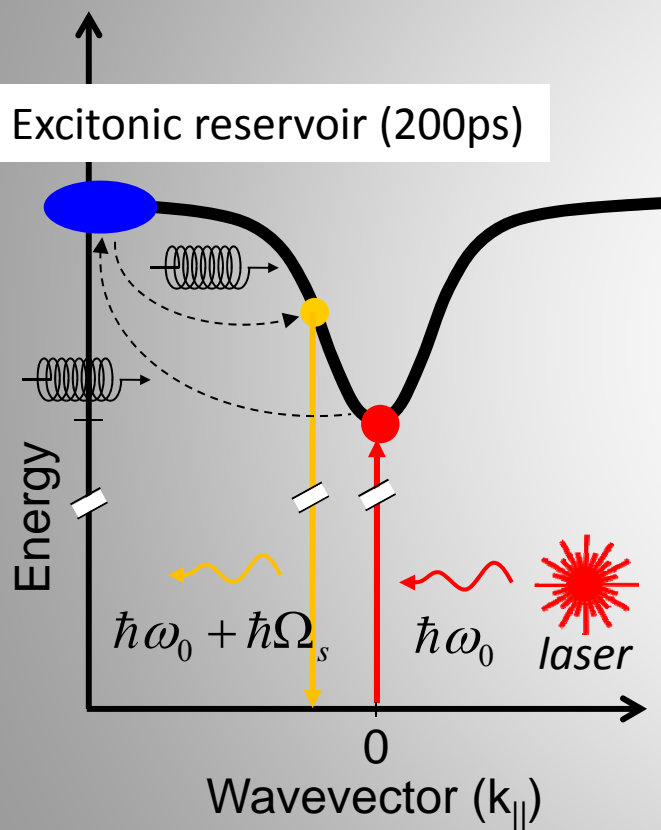
# anti-Stokes fluorescence of polaritons





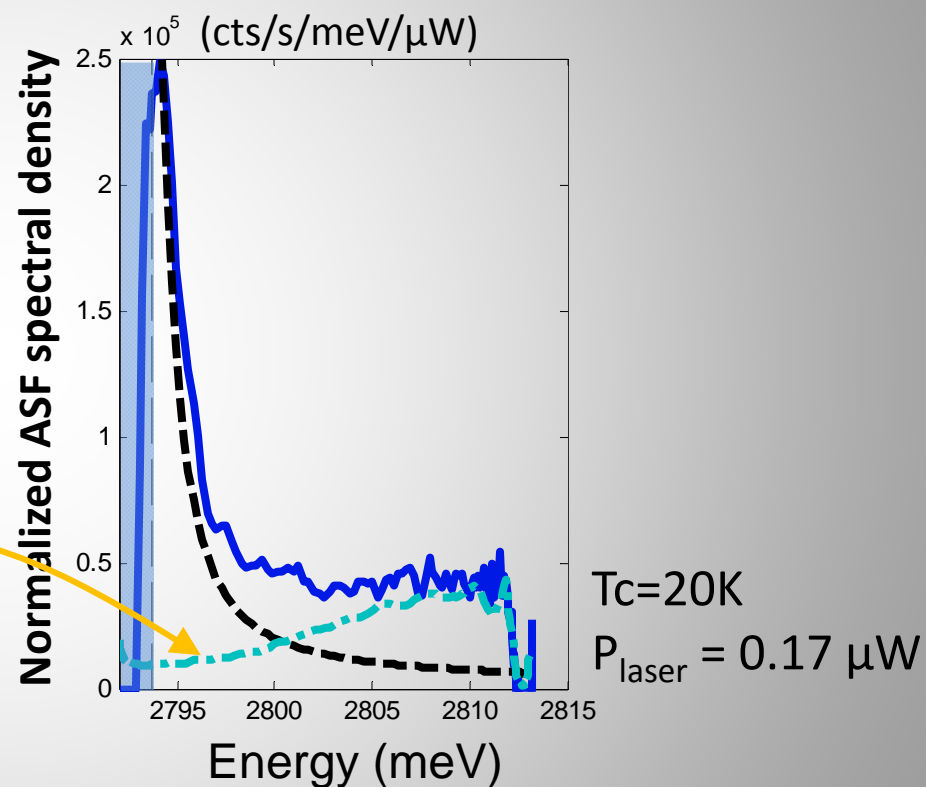
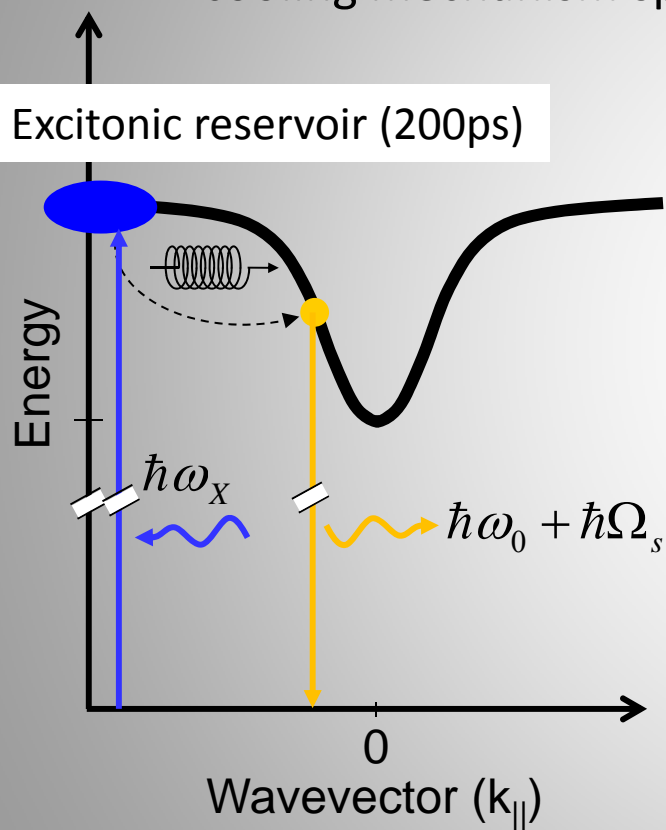
# anti-Stokes fluorescence of polaritons

Slow cooling mechanism ❄️



# anti-Stokes fluorescence of polaritons

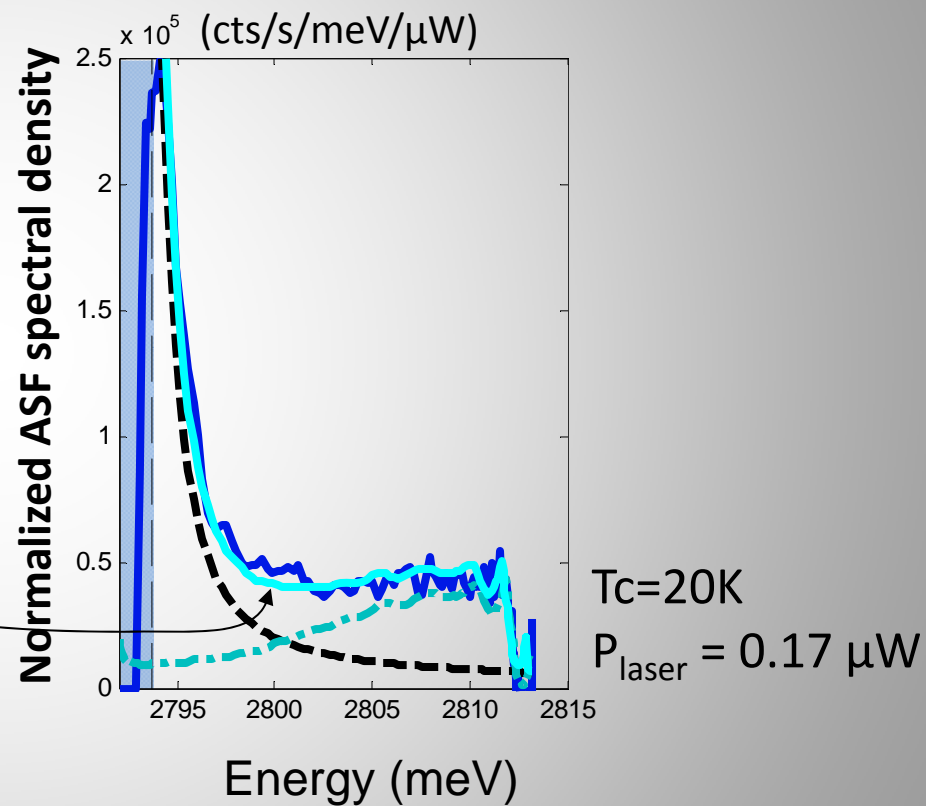
Measurement of the **Slow** cooling mechanism spectrum



# anti-Stokes fluorescence of polaritons



**Fast + Slow** cooling mechanisms



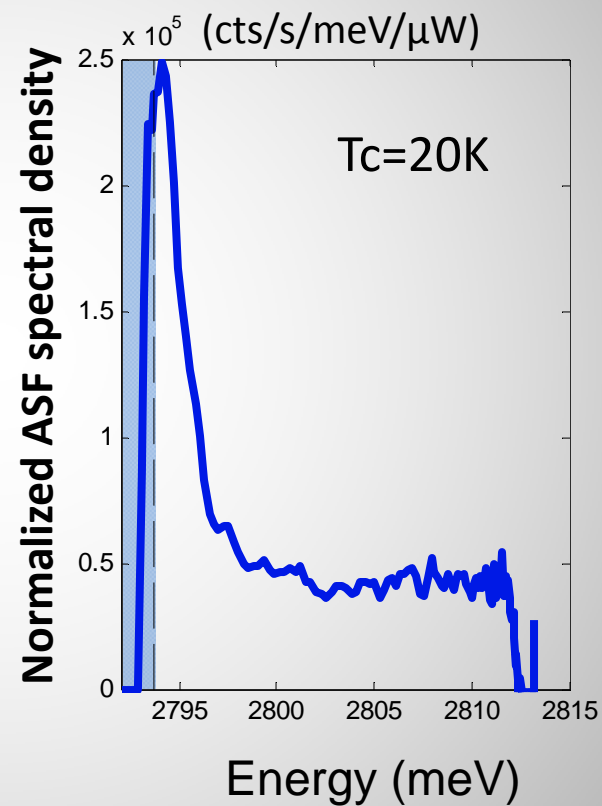
# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

Laser power dependence

$$\propto P_{las}$$

$$P_{laser} = 0.17 \mu\text{W}$$



# anti-Stokes fluorescence of polaritons

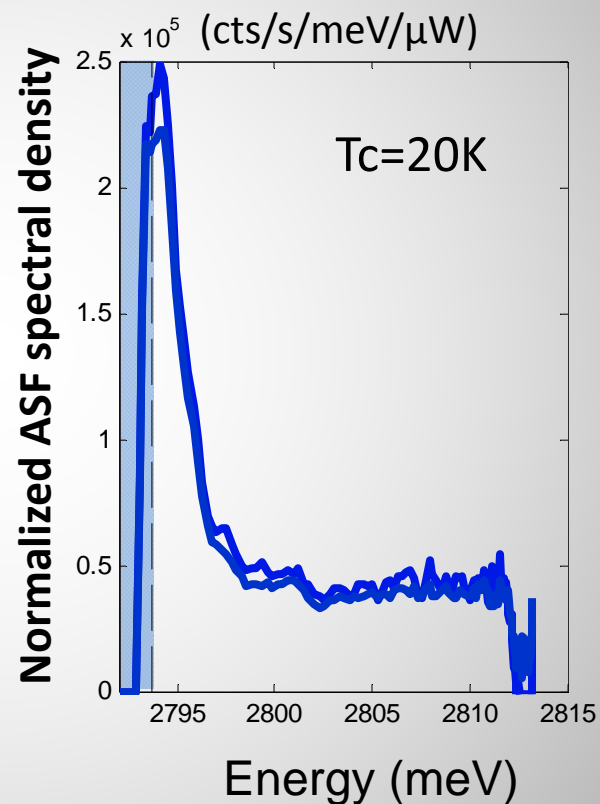
$$I_{las} / P_{las}$$

Laser power dependence

$$\propto P_{las}$$

$$P_{laser} = 0.31 \mu\text{W}$$

$$P_{laser} = 0.17 \mu\text{W}$$



# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

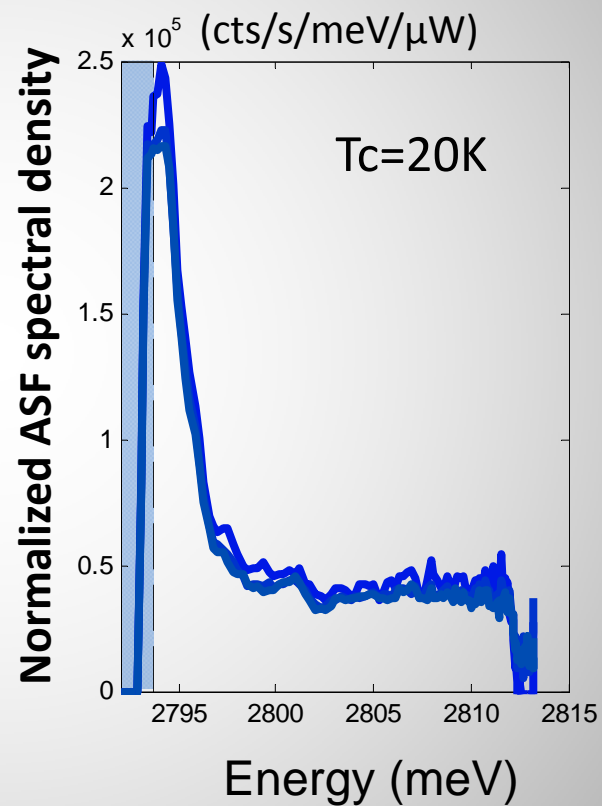
Laser power dependence

$$\propto P_{las}$$

$$P_{laser} = 0.48 \mu\text{W}$$

$$P_{laser} = 0.31 \mu\text{W}$$

$$P_{laser} = 0.17 \mu\text{W}$$



# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

Laser power dependence

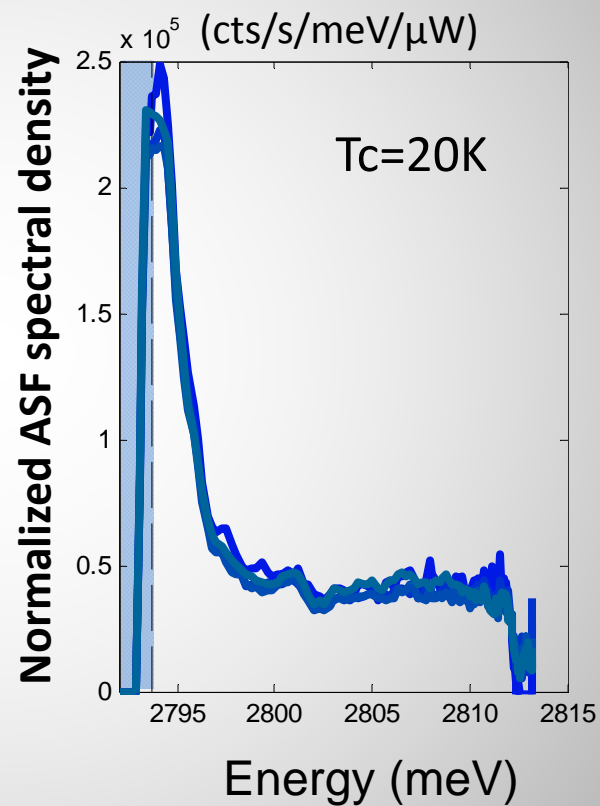
$$\propto P_{las}$$

$$P_{laser} = 1.26 \mu\text{W}$$

$$P_{laser} = 0.48 \mu\text{W}$$

$$P_{laser} = 0.31 \mu\text{W}$$

$$P_{laser} = 0.17 \mu\text{W}$$



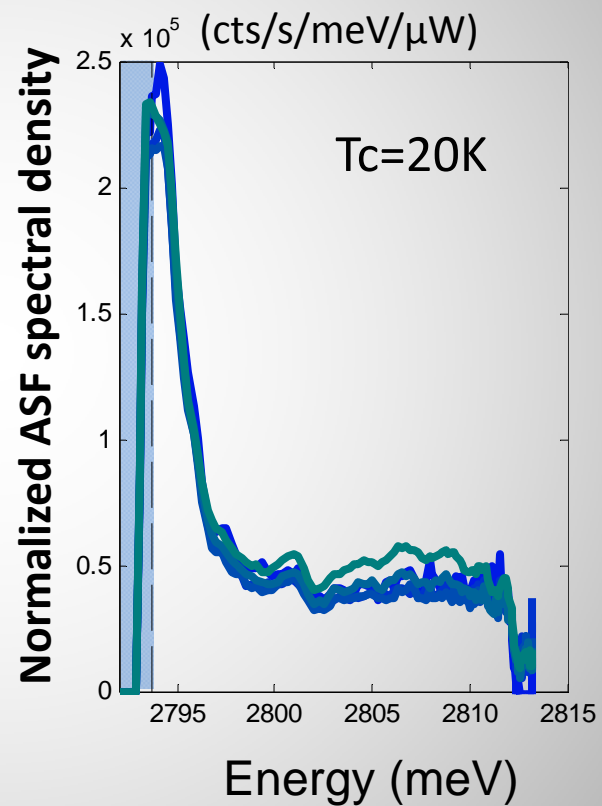
# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

Laser power dependence

$$\propto P_{las}^2$$

$$P_{laser} = 3.30 \mu\text{W}$$





# anti-Stokes fluorescence of polaritons

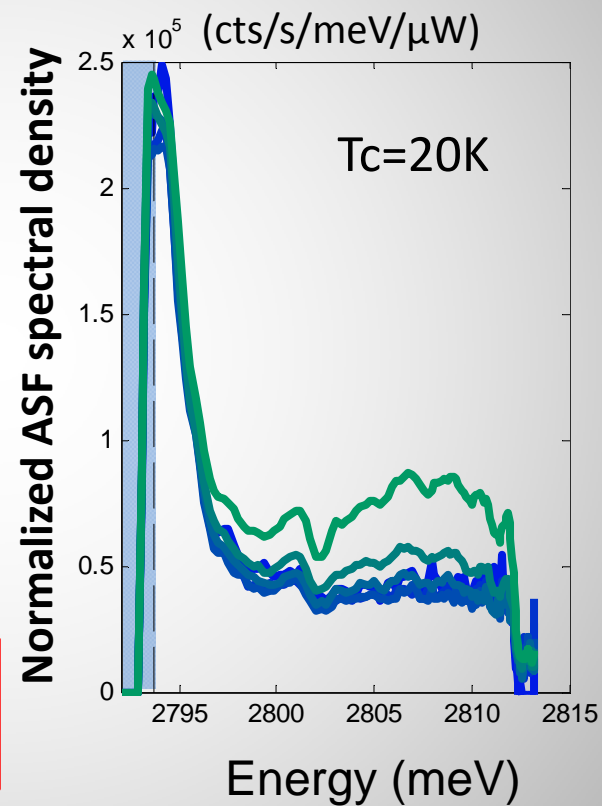
$$I_{las} / P_{las}$$

Laser power dependence

$$\propto P_{las}^2$$

$$P_{laser} = 3.30 \mu\text{W}$$

$$P_{laser} = 11.16 \mu\text{W}$$



# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

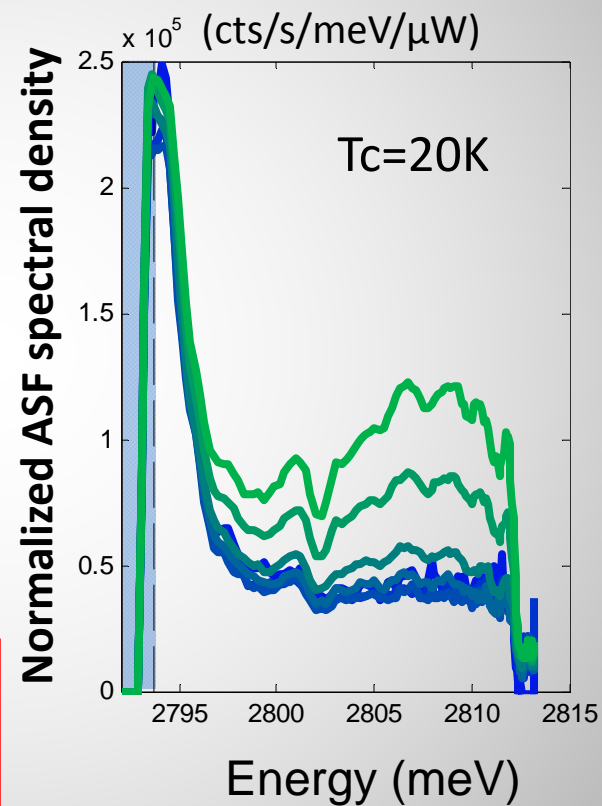
Laser power dependence

$$\propto P_{las}^2$$

$$P_{laser} = 3.30 \mu\text{W}$$

$$P_{laser} = 11.16 \mu\text{W}$$

$$P_{laser} = 26.20 \mu\text{W}$$



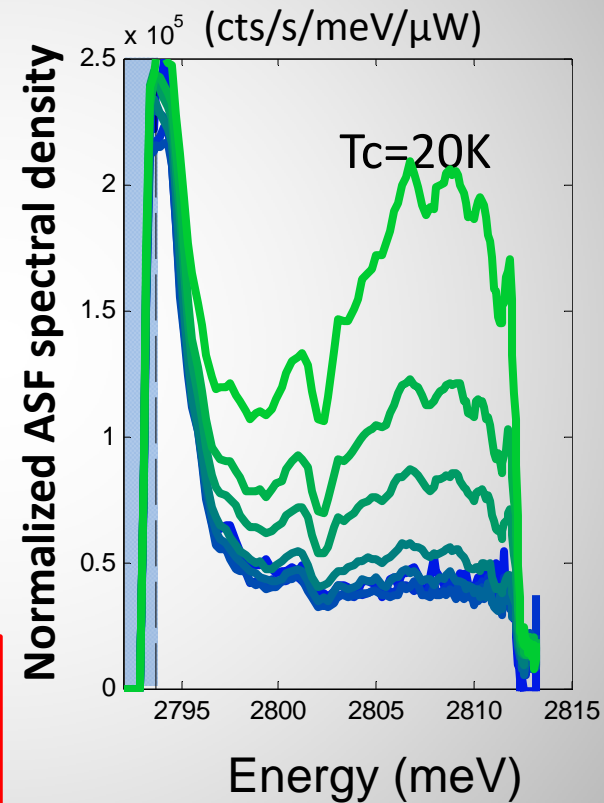
# anti-Stokes fluorescence of polaritons

$$I_{las} / P_{las}$$

Laser power dependence

$$\propto P_{las}^2$$

$P_{laser} = 3.30 \mu\text{W}$   
 $P_{laser} = 11.16 \mu\text{W}$   
 $P_{laser} = 26.20 \mu\text{W}$   
 $P_{laser} = 83.71 \mu\text{W}$

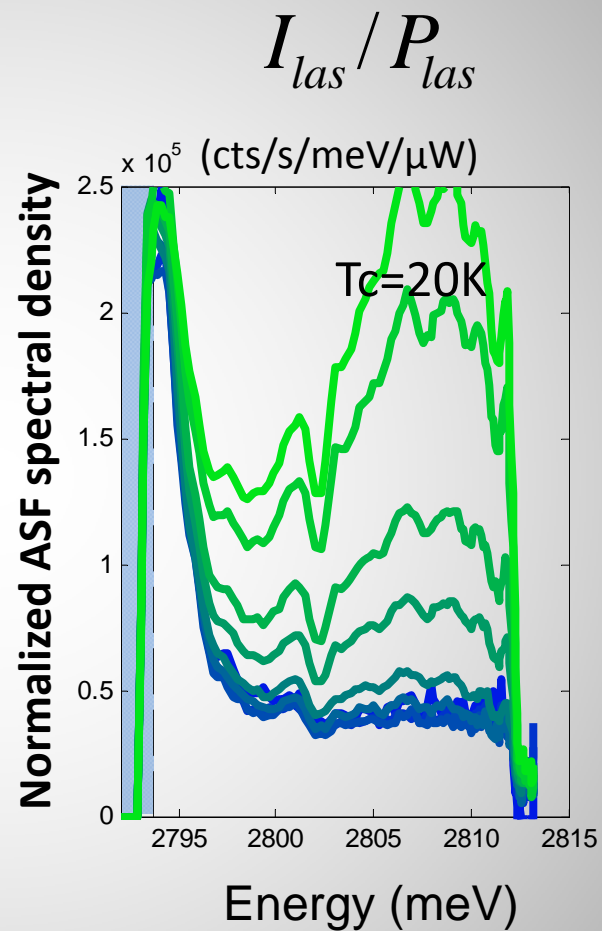


# anti-Stokes fluorescence of polaritons

Laser power dependence

$$\propto P_{las}^2$$

$P_{laser} = 3.30 \mu\text{W}$   
 $P_{laser} = 11.16 \mu\text{W}$   
 $P_{laser} = 26.20 \mu\text{W}$   
 $P_{laser} = 83.71 \mu\text{W}$   
 $P_{laser} = 196.53 \mu\text{W}$



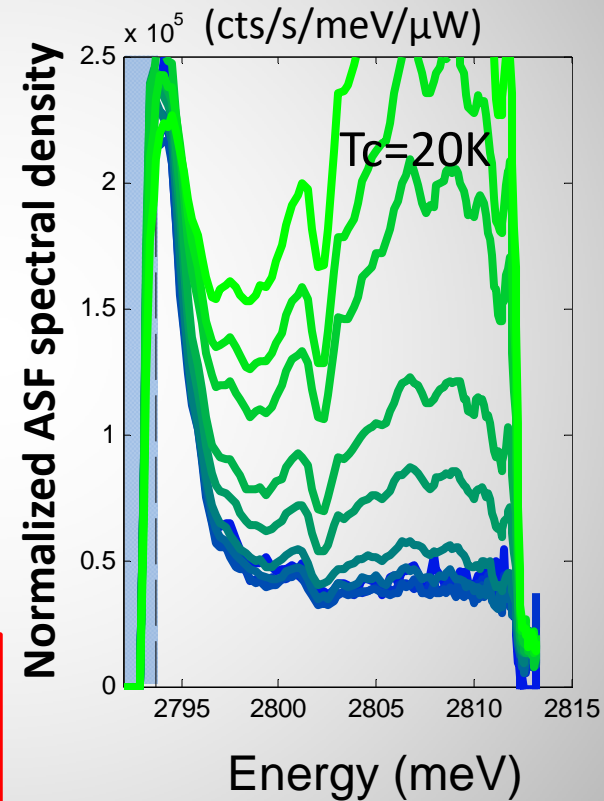
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$$I_{las} / P_{las}$$

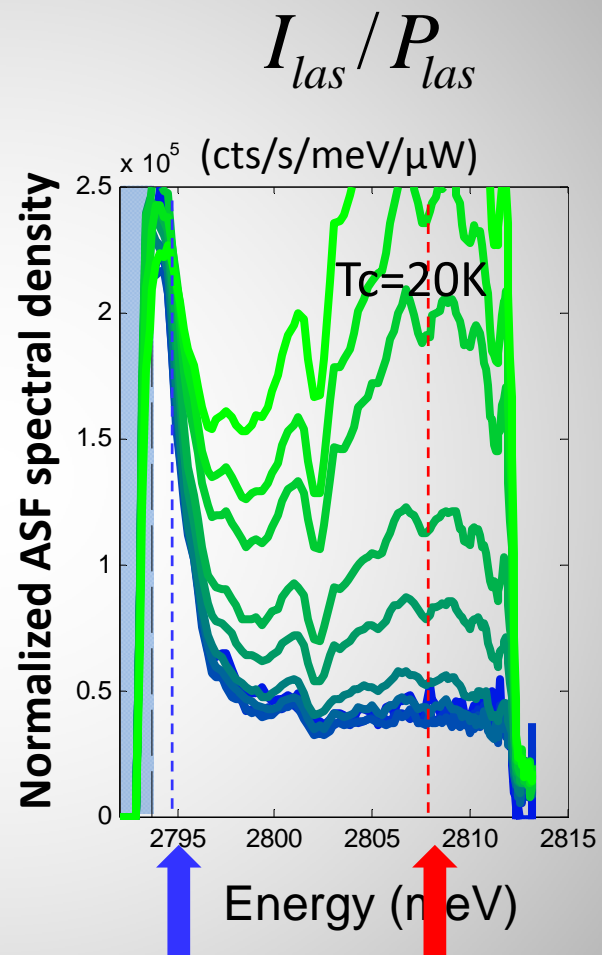
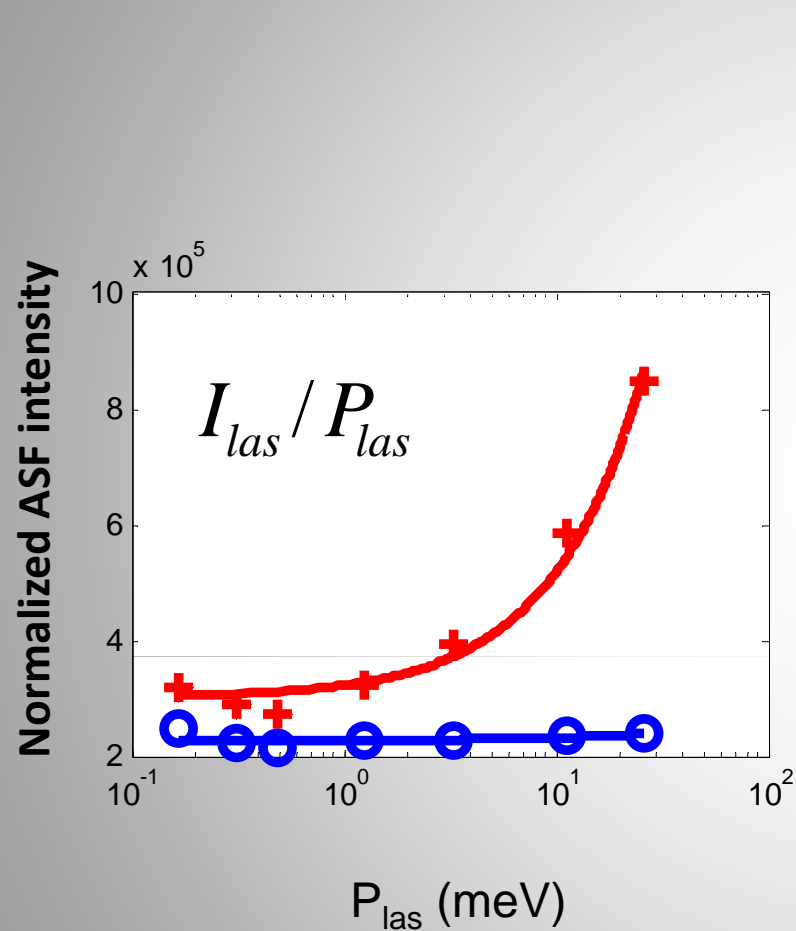
Laser power dependence

$$\propto P_{las}^2$$

$P_{laser} = 3.30 \mu\text{W}$   
 $P_{laser} = 11.16 \mu\text{W}$   
 $P_{laser} = 26.20 \mu\text{W}$   
 $P_{laser} = 83.71 \mu\text{W}$   
 $P_{laser} = 196.53 \mu\text{W}$   
 $P_{laser} = 500.00 \mu\text{W}$

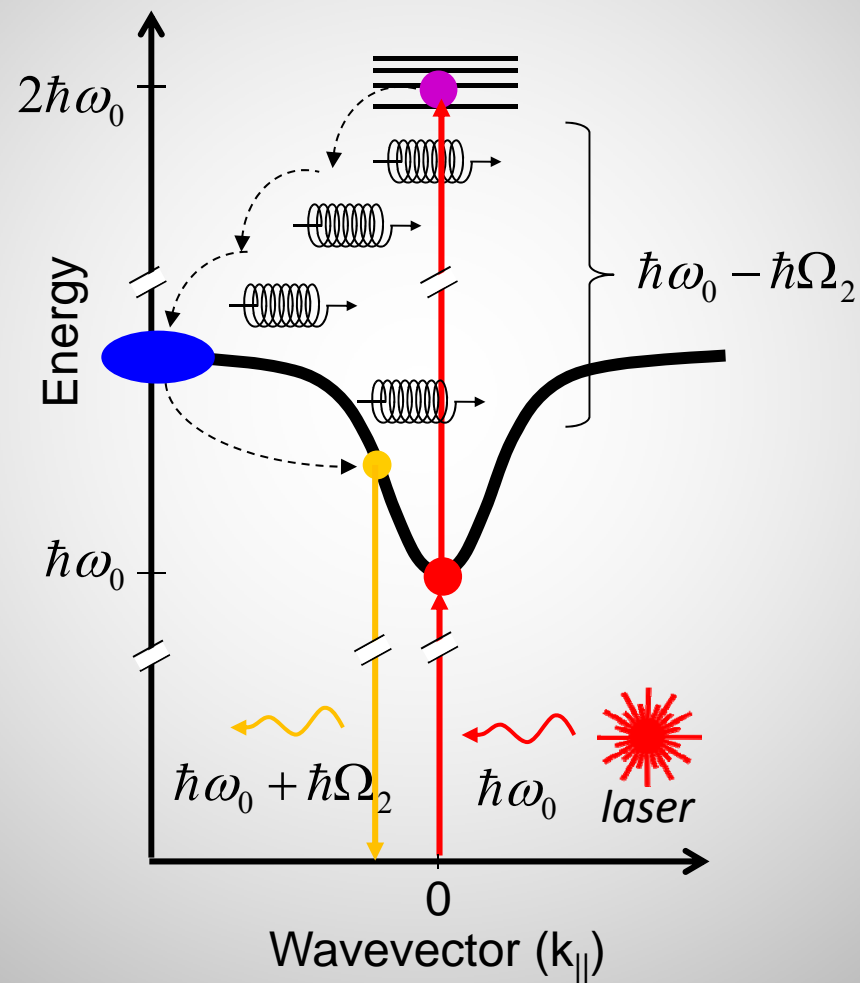


# anti-Stokes fluorescence of polaritons



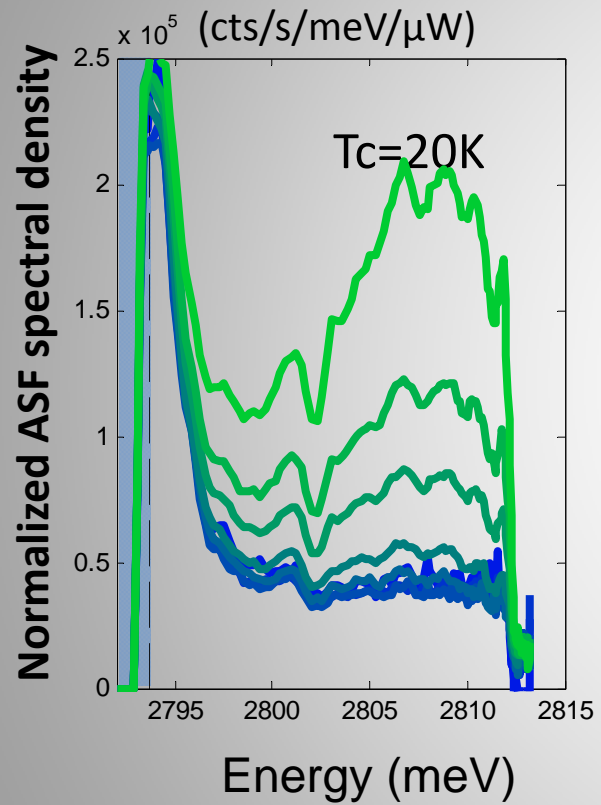
# anti-Stokes fluorescence of polaritons

2-photon absorption heating mechanism



## Intermediate summary

$$I_{las} / P_{las}$$

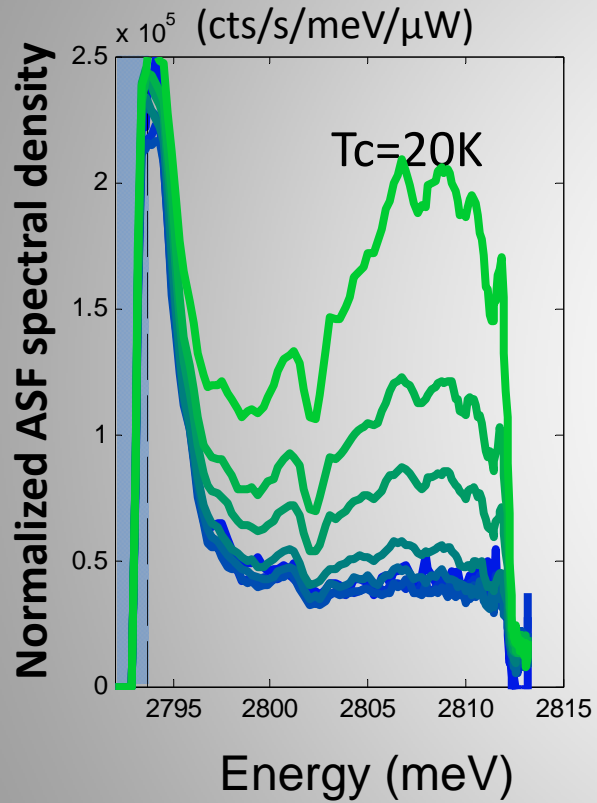


+ Setup detection efficiency calibration  $\eta=1.1\%$



# Intermediate summary

$$I_{las} / P_{las}$$

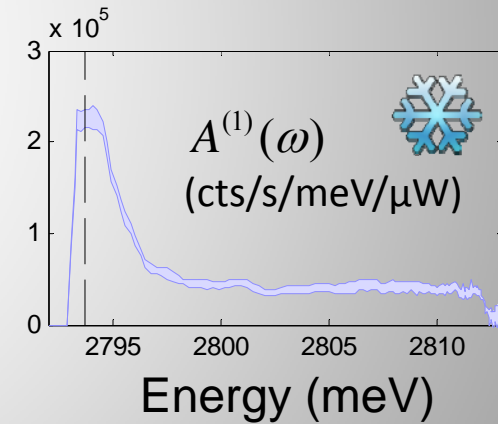


$\propto P_{las}$

**Slow** cooling mechanism

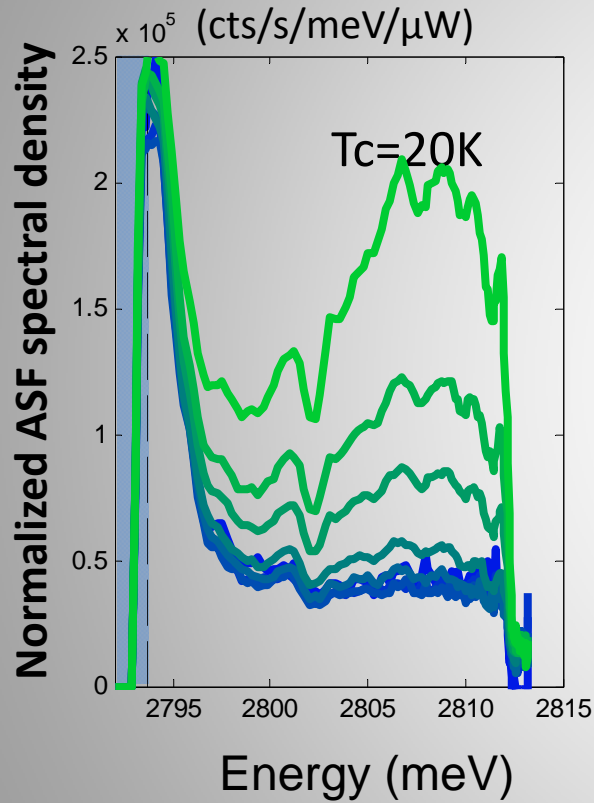


**Fast** cooling mechanism



# Intermediate summary

$$I_{las} / P_{las}$$

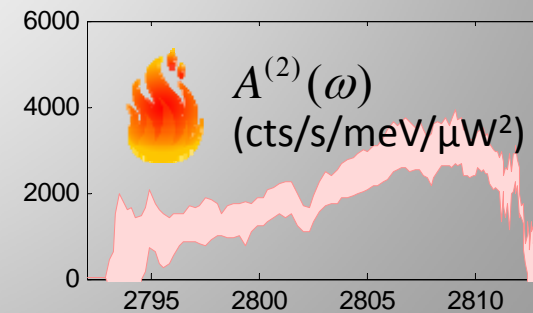
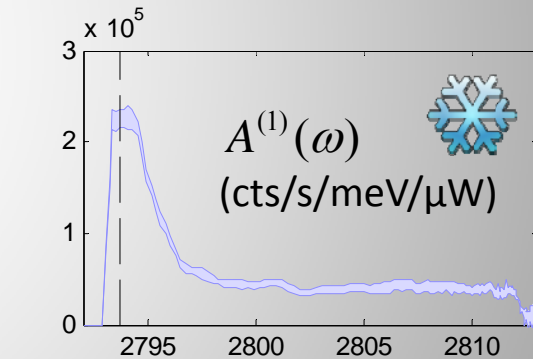


$$\propto P_{las}$$

Slow cooling mechanism



Fast cooling mechanism



$$\propto P_{las}^2$$

2-photon abs. heating

# Quantitative thermal Balance

Cooling power (Watts)

Absorbed phonon energy

**Fast and slow** cooling event rate



$$P_{\text{fr}} = \int d\omega \left\{ P_{\text{las}} \hbar(\omega - \omega_0) A^{(1)}(\omega) - P_{\text{las}}^2 \hbar(2\omega_0 - \omega) A^{(2)}(\omega) \right\}$$


# Quantitative thermal Balance

Cooling power (Watts)

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2-photon absorption rate 

phonon cascade emission energy

# Quantitative thermal Balance

Cooling power (Watts)

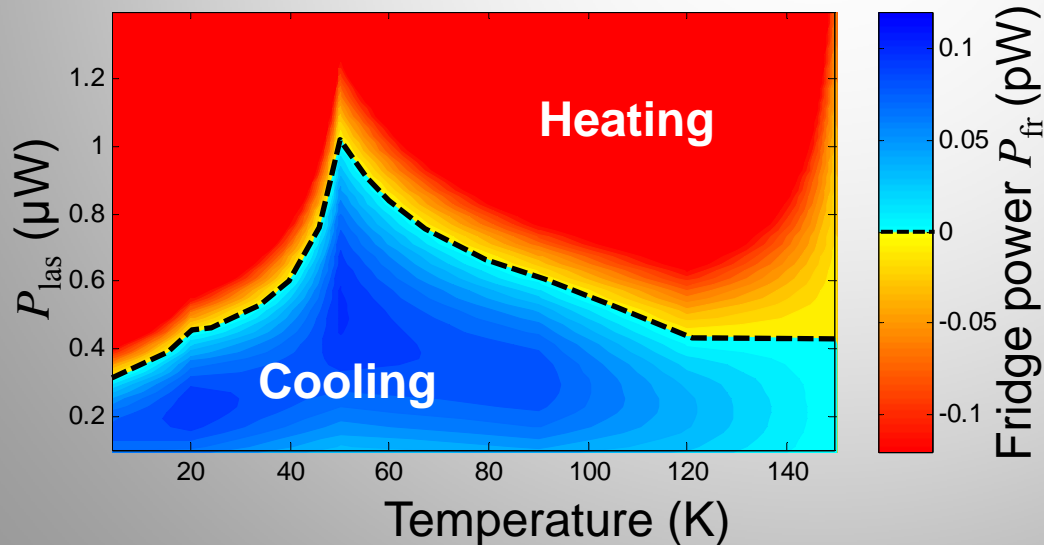
Absorbed phonon energy

Fast and slow cooling event rate ❄️

$$P_{fr} = \int d\omega \left\{ P_{las} \hbar(\omega - \omega_0) A^{(1)}(\omega) - P_{las}^2 \hbar(2\omega_0 - \omega) A^{(2)}(\omega) \right\}$$

2-photon absorption rate 🔥

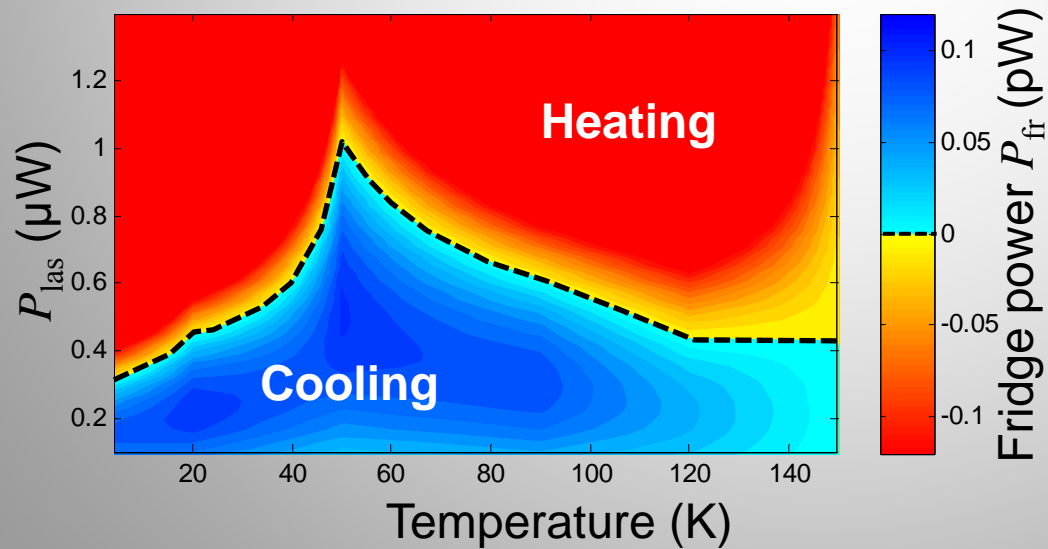
phonon cascade emission energy



- $P_{fr, \max} = 0.1 \pm 0.02$  pW
- $p_{fr, \max} = 80 \pm 16 \mu\text{W}/\text{cm}^3$

# Quantitative thermal Balance

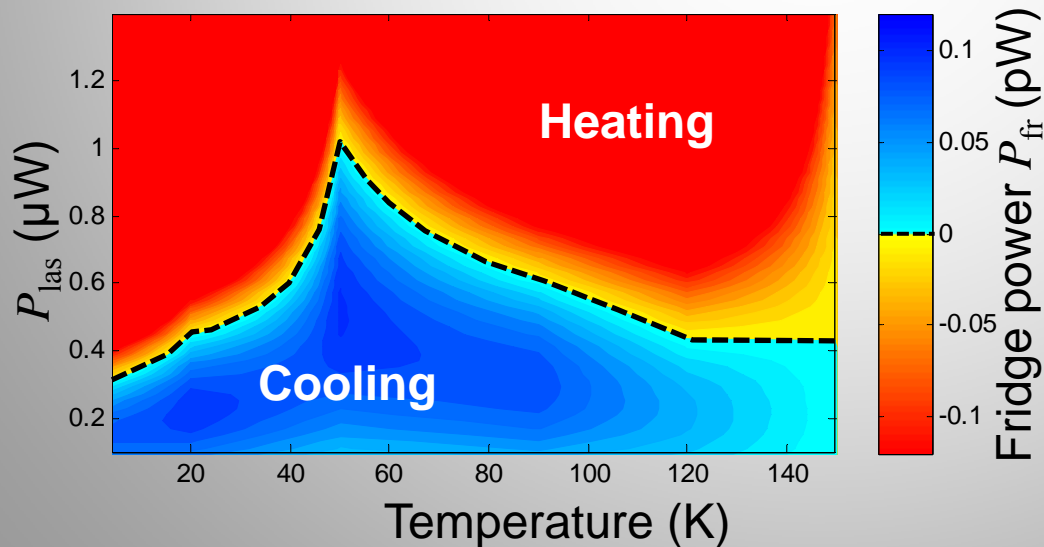
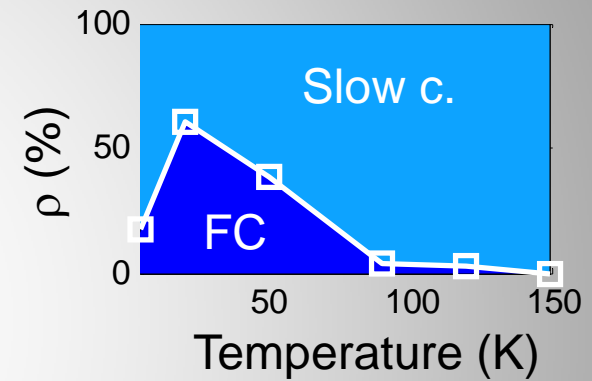
- No temperature cutoff



- $P_{\text{fr, max}} = 0.1 \pm 0.02 \text{ pW}$
- $p_{\text{fr, max}} = 80 \pm 16 \mu\text{W}/\text{cm}^3$

# Quantitative thermal Balance

- No temperature cutoff
- High participation ratio  $\rho$  of **fast** cooling mechanism

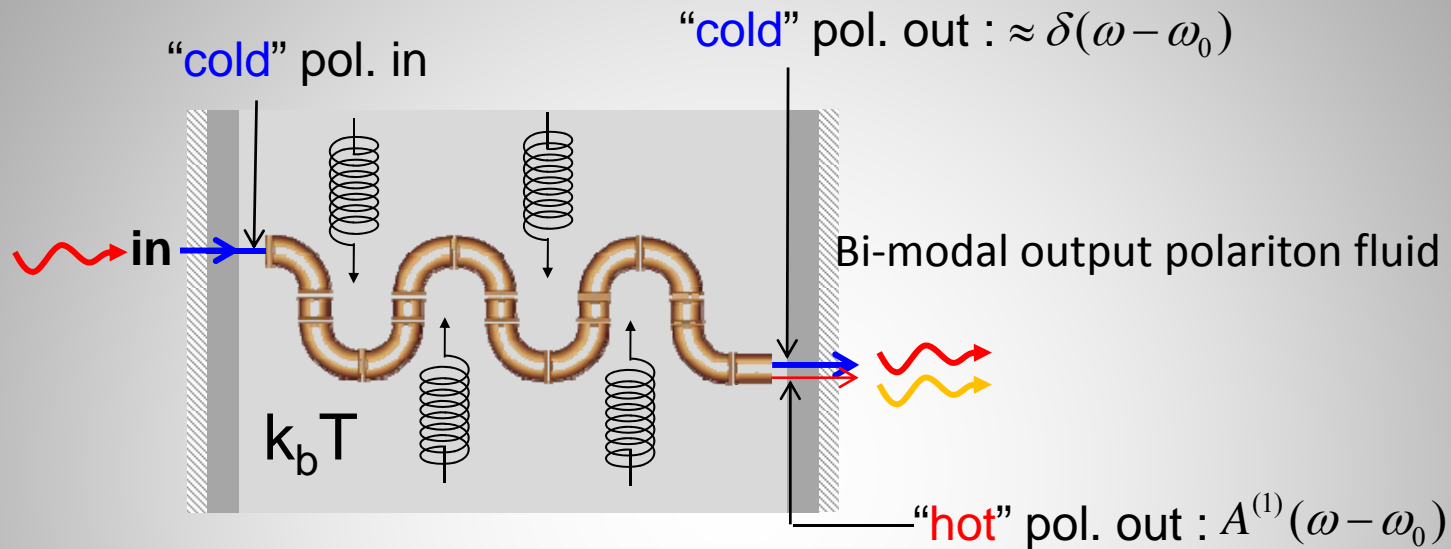


- $P_{fr, \max} = 0.1 \pm 0.02$  pW
- $p_{fr, \max} = 80 \pm 16 \mu$ W/cm<sup>3</sup>

What does the heated polariton gas look like?



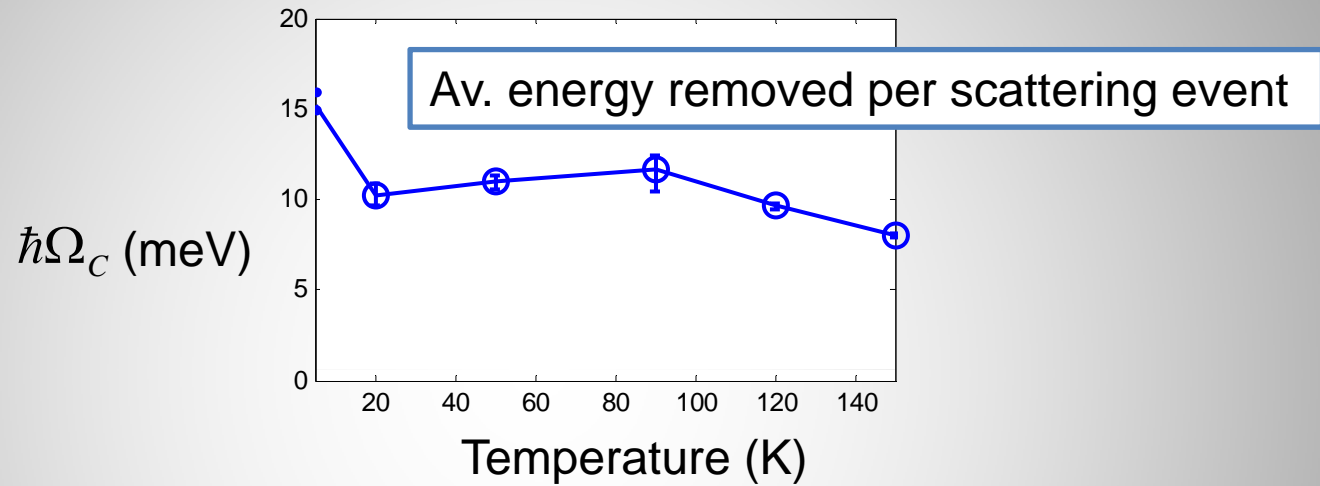
## What does the heated polariton gas looks like?



- No internal equilibration (vanishing pp interaction regime)  
→ Bi-modale polariton distribution : a « cold » and a « hot » fluid coexist

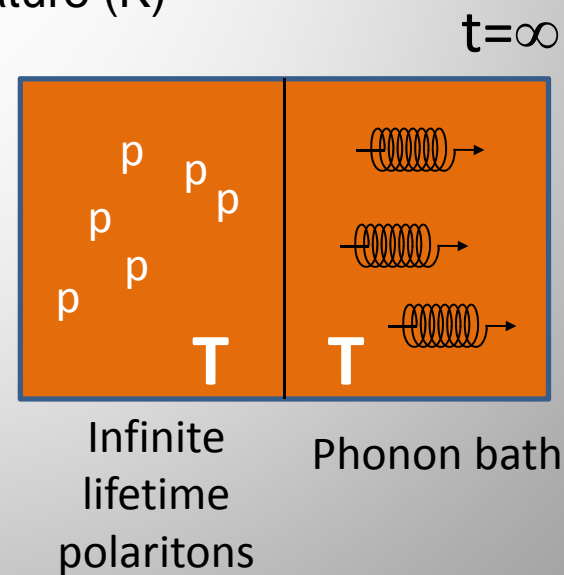
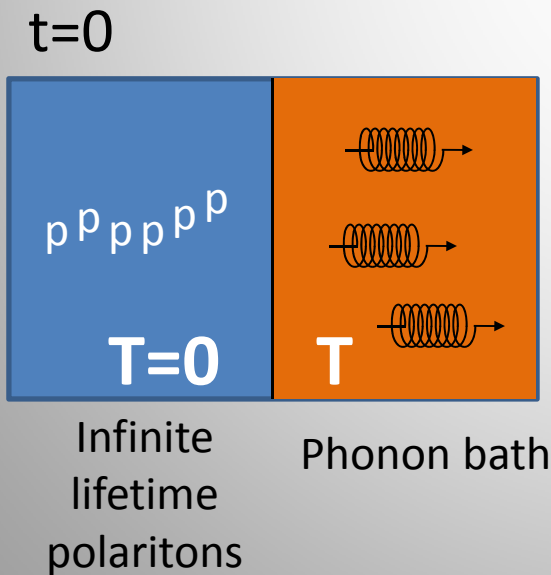
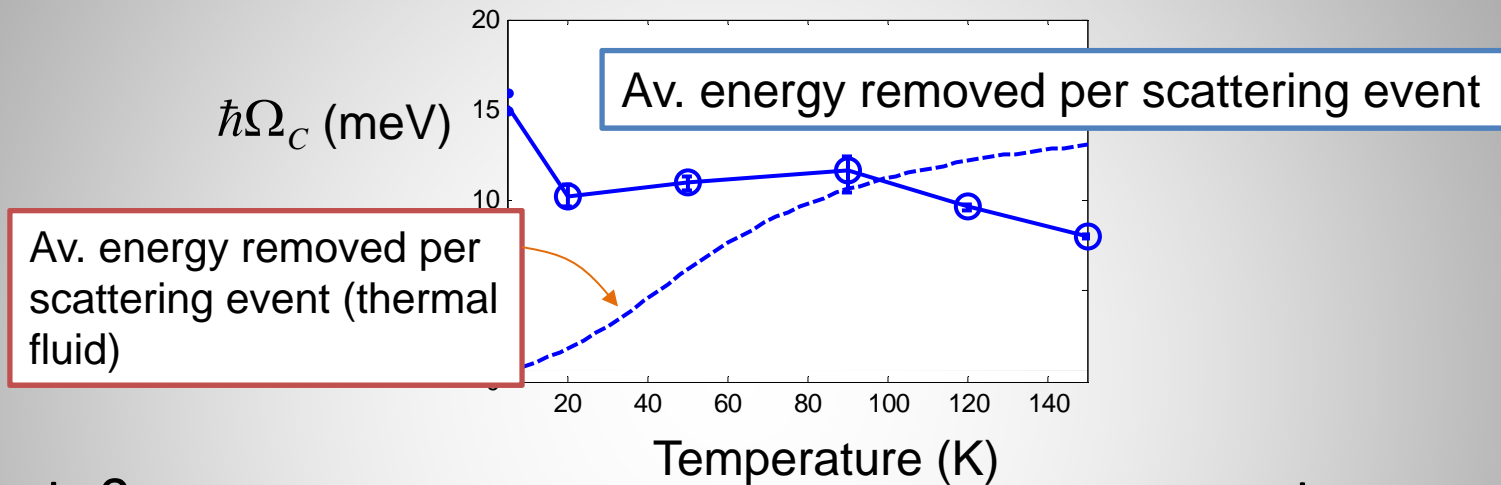
# What does the heated polariton gas looks like?

« hot » polariton fluid properties = polariton that did interact with thermal phonons



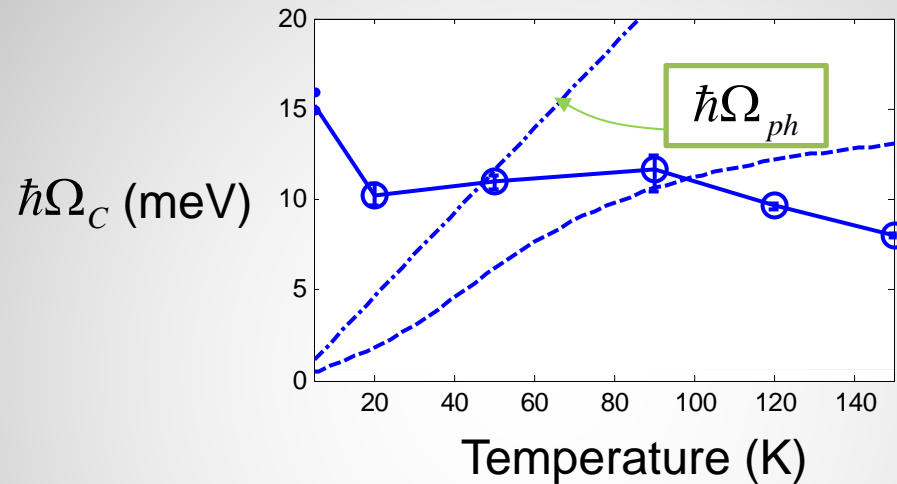
# What does the heated polariton gas looks like?

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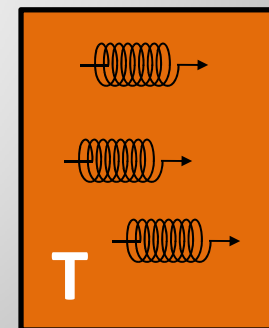


# What does the heated polariton gas looks like?

« hot » polariton fluid properties = polariton that did interact with thermal phonons



Heated polaritons are “hotter” than the phonons themselves !



Phonon bath

## Conclusion

### Properties of a polaritonic refrigerent

- Net Positive cooling power at low laser power
- Involves an ultrafast cooling dynamics mechanism (1ps)
- No temperature cutoff
- Full optical access to thermodynamical properties

- Main limitation so far : 2-photon absorption

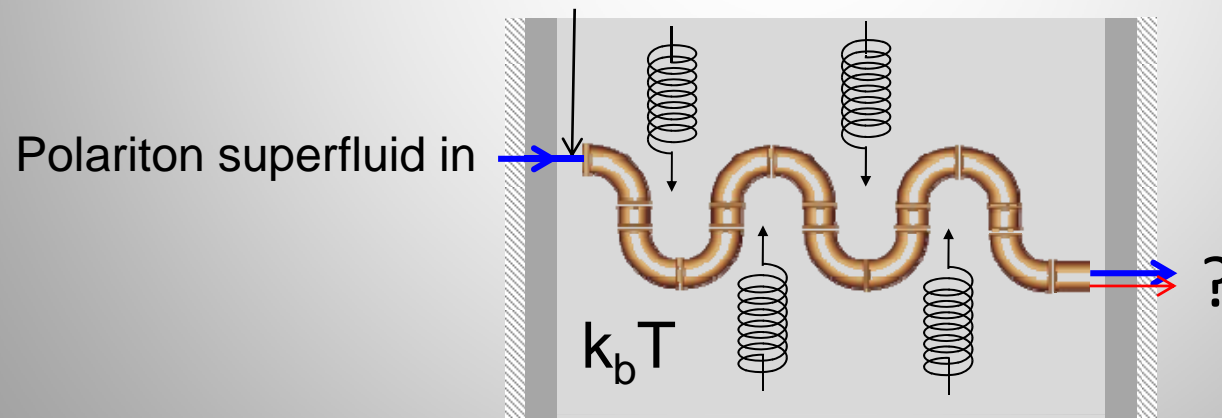
### « Cold » injected polaritons behave like an out-of-equilibrium refrigerant fluid

- Bi-modal « cold » and « hot » fluid
- at low T, polaritons removes thermal phonons of higher energy than normally allowed by thermal equilibrium
- non-eq. can be a resource !

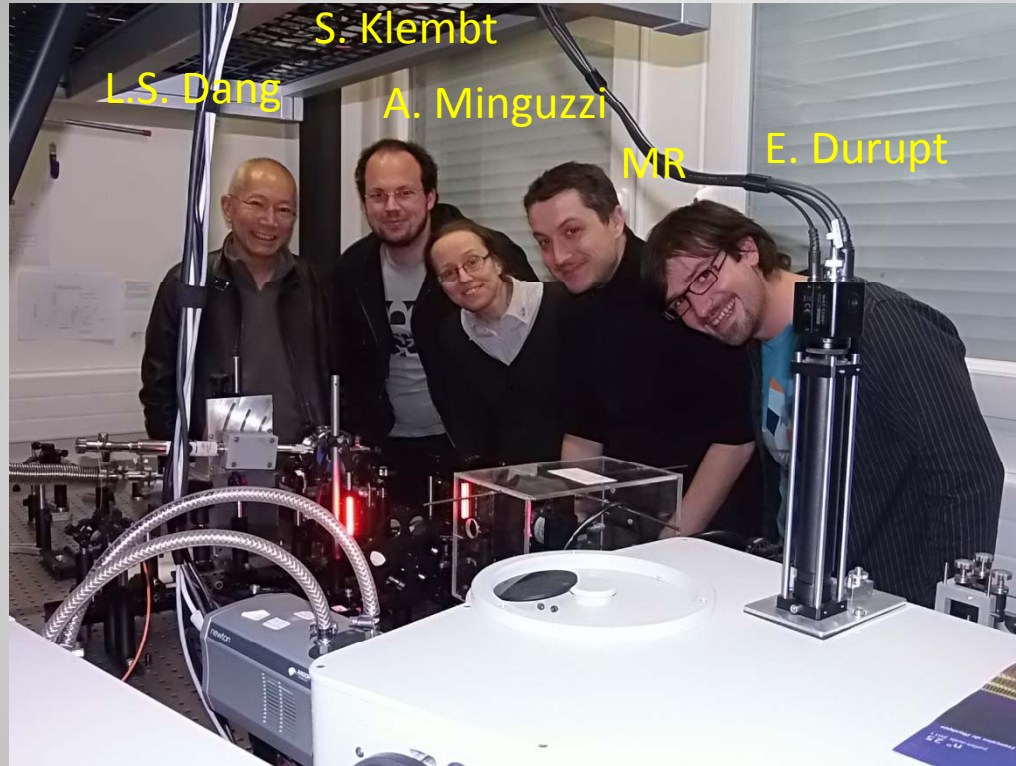
## Outlook

### **Non thermal character is fully tunable ! :**

- pp interactions  $\Leftrightarrow$  internal equilibrations
- A tunable thermal reservoir can be added : externally pumped excitons
- **Thermodynamical properties of polariton superfluids**  
= thermodynamics of a (out-of-eq.) weakly interacting Bose gas exchanging heat with a thermal reservoir



# Acknowledgements



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A. Baas

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