



# On the mechanism of Non-Photochemical Laser Induced Nucleation

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## NPLIN = Non-Photochemical Laser Induced Nucleation

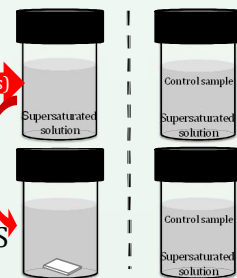
Nucleation process discovered by Garetz *et al*<sup>1</sup> (1996)

- NPLIN allows :
- A temporal control of nucleation ( $\sim$  induction time)
  - An increase of the probability of nucleation
  - A control of the number of crystals

What is the mechanism of this phenomenon?

Different hypotheses have been investigated through supersaturated aqueous solutions of  $K_2SO_4$  or EDS

### Cavitation, Optical Kerr Effect and nanoparticle heating



### Cavitation : bubble generation<sup>2</sup>

**Hypothesis :**

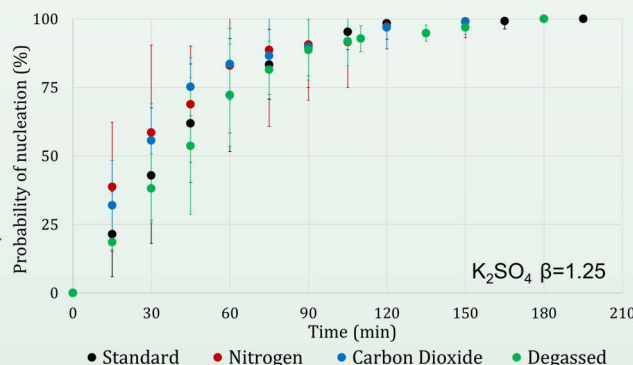


If bubbles were composed of dissolved gas

→ Different gas should lead to different NPLIN kinetics

**Experiment :** Bubbling of gas to control composition and nature of gases in supersaturated aqueous solutions of  $K_2SO_4$

- NPLIN behavior is similar for each gas condition,  
There is no influence of gases

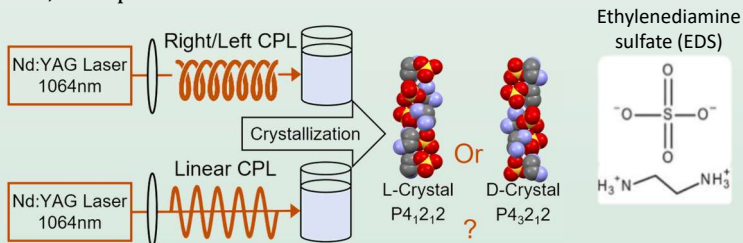


### Can NPLIN be a stereoselective process?

**Hypothesis :** OKE (Optical Kerr Effect<sup>1</sup>)

If an alignment of the dipoles along the electromagnetic field occurs

Then, laser polarization can influence NPLIN



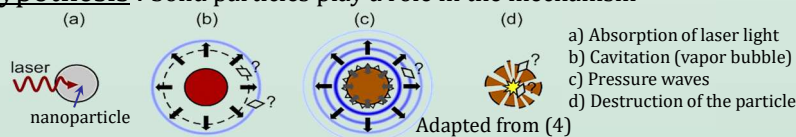
### Results: EDS $\beta=1.10$

Polarization	$N_{vial}$	$N_{crystal}$	$N_D$	$N_L$	$Cee \pm \sigma$
Left Circularly Polarized Light	89	194	99	95	$0.02 \pm 0.07$
Right Circularly Polarized Light	86	190	81	109	$-0.14 \pm 0.07$
Linearly Polarized Light	91	251	129	122	$0.03 \pm 0.06$

→ Crystal Enantiomeric Excess (Cee) shows no significant change upon changing laser polarization NPLIN is non-stereoselective. OKE is not applicable to EDS.

### Nanoparticle heating<sup>3</sup>

**Hypothesis :** Solid particles play a role in the mechanism

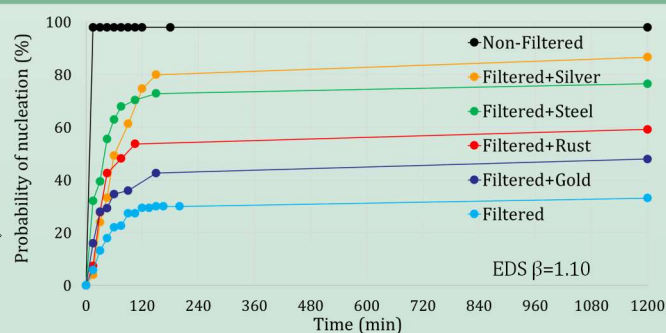


**Experiment :** Filtration and doping of supersaturated solution

→ ICP analysis of removed impurities

Concentration (mg/L)	Fe	Ni	Cr	S
Control	3.3	0.28	0.06	0.14
Impurity Sample	8.2	0.76	0.71	1.60

→ Stainless steel Composition



→ Filtration decreases the probability of nucleation, but addition of different particle lead to different kinetics behavior

### Conclusions

Laser polarization geometry has no incidence on nucleation :

→ OKE mechanism is invalid for EDS

If there is Cavitation, bubbles might not be composed of gas

→ 2<sup>nd</sup> hypothesis bubbles might be composed of solvent vapor

Nanoparticle heating seems the best hypothesis since presence or absence of particles has a strong influence on NPLIN behavior but it is not clear how particles are involved in the nucleation of the crystal (Hypothesis b, c or d from(4)).

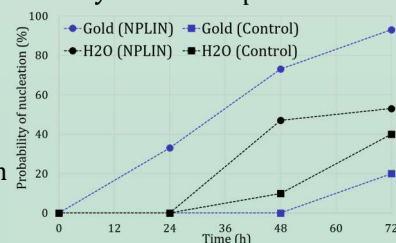
### Perspectives

Possibility to enhance the NPLIN effect by addition of particles.

For Diprophylline/  $H_2O$   $\beta=3.25$   
Doping with Gold nanoparticles

- $\sim$  Induction time
- $\nearrow$  Probability of nucleation

Different particles concentration and nature have to be tested



### References

- (1) Garetz, B. A.; Aber, J. E.; Goddard, N. L.; Young, R. G.; Myerson, A. S. *Physical review letters* 1996, 77 (16), 3475 (2) Soare, A.; Dijkink, R.; Pascual, M. R.; Sun, C.; Cains, P. W.; Lohse, D.; Stankiewicz, A. I.; Kramer, H. J. M. *Crystal Growth & Design* 2011, 11 (6), 2311–2316. (3) Ward, M. R.; Mackenzie, A. M.; Alexander, A. J. *Crystal Growth & Design* 2016, 16 (12), 6790–6796. (4) Alexander, A. J.; Camp, P. J. *J. Chem. Phys.* 2019, 150 (4), 040901.