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On the mechanism of Non-Photochemical Laser Induced Nucleation

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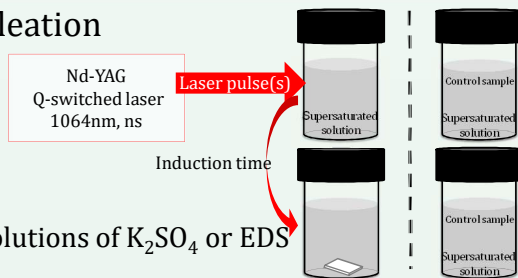
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NPLIN = Non-Photochemical Laser Induced Nucleation

Nucleation process discovered by Garetz *et al*¹ (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 generation²

Hypothesis :

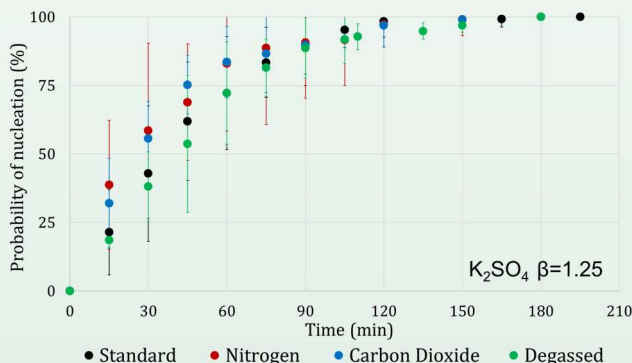


If bubbles were composed of dissolved gas

↳ 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

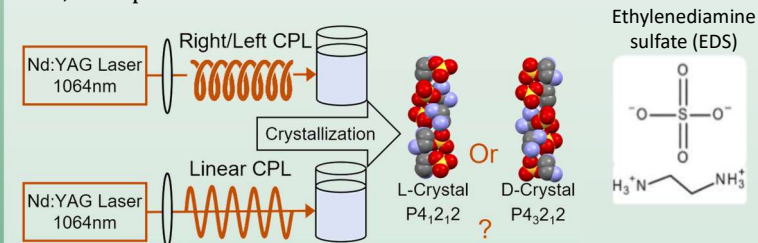


Can NPLIN be a stereoselective process?

Hypothesis : OKE (Optical Kerr Effect)¹

If an alignment of the dipoles along the electromagnetic field occurs

Then, laser polarization can influence NPLIN



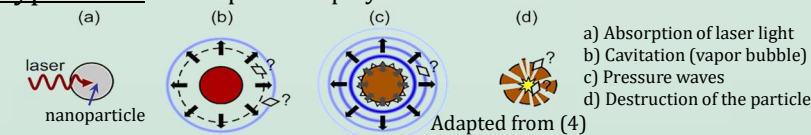
Results: EDS beta=1.10

Polarization	N _{vial}	N _{crystal}	N _D	N _L	Cee±σ
Left Circularly Polarized Light	89	194	99	95	0.02±0.07
Right Circularly Polarized Light	86	190	81	109	-0.14±0.07
Linearly Polarized Light	91	251	129	122	0.03±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 heating³

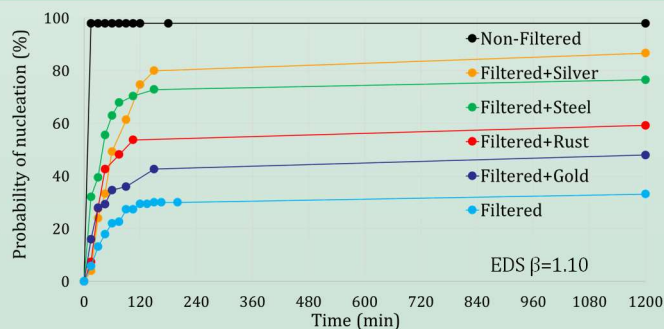
Hypothesis : Solid particles play a role in the mechanism



Experiment : Filtration and doping of supersaturated solution

↳ ICP analysis of removed impurities

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



→ 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

↳ 2nd hypothesis bubbles might be composed of solvent vapor

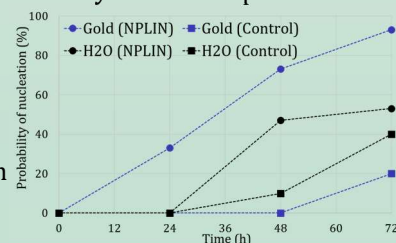
Nanoparticle heating seems the best hypothesis since presence or absence of particles has a strong influence on NPLIN behavior but it is not clear how particles are involved in the nucleation of the crystal (Hypothesis b, c or d from(4)).

Perspectives

Possibility to enhance the NPLIN effect by addition of particles.

- For Diprophylline/ H₂O 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.