



HAL
open science

On the mechanism of Non-Photochemical Laser Induced Nucleation

Melody Briard, Clément Brandel, Gérard Coquerel, Valerie Dupray

► **To cite this version:**

Melody Briard, Clément Brandel, Gérard Coquerel, Valerie Dupray. On the mechanism of Non-Photochemical Laser Induced Nucleation. CRYSTAL GROWTH OF ORGANIC MATERIALS - CGOM14, Sep 2022, Brussels, Belgium. hal-03777825

HAL Id: hal-03777825

<https://normandie-univ.hal.science/hal-03777825>

Submitted on 15 Sep 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Normandie Université

Mélody Briard¹, Clément Brandel¹, Gérard Coquerel¹, Valérie Dupray¹

¹ Laboratoire SMS, UR 3233, Université de Rouen-Normandie, 76821 Mont Saint Aignan

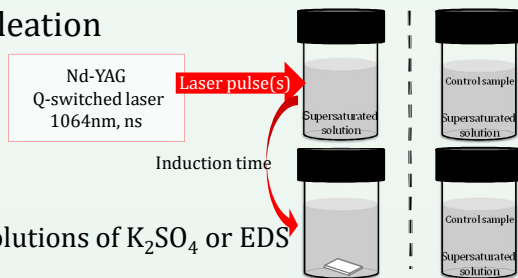
melody.briard1@univ-rouen.fr



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 :

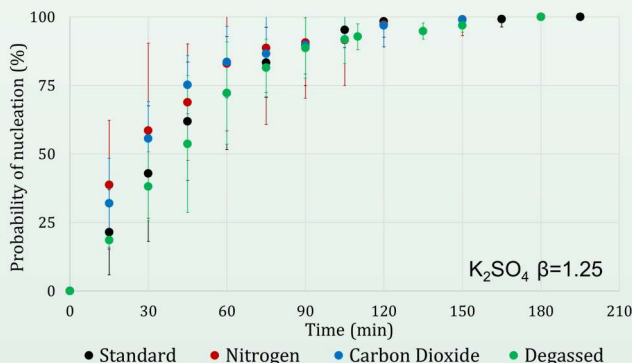


If bubbles were composed of dissolved gas

↳ 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

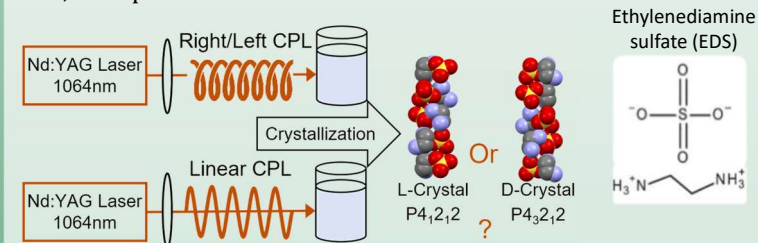


Can NPLIN be a stereoselective process?

Hypothesis : OKE (Optical Kerr Effect)¹

If an alignment of the dipoles along the electromagnetic field occurs

Then, laser polarization can influence NPLIN



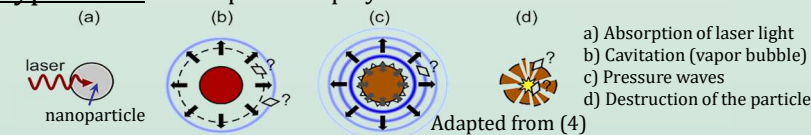
Results: EDS $\beta=1.10$

Polarization	N_{vial}	$N_{crystal}$	N_D	N_L	$Cee \pm \sigma$
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

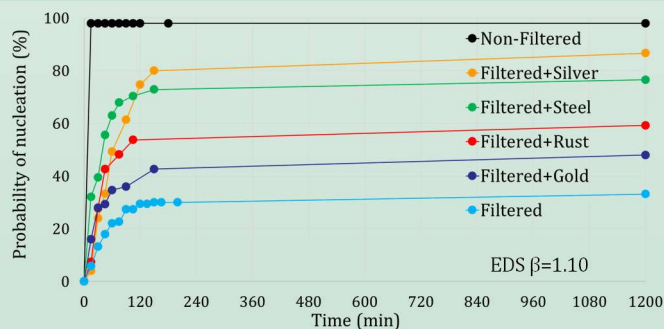
Hypothesis : Solid particles play a role in the mechanism



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

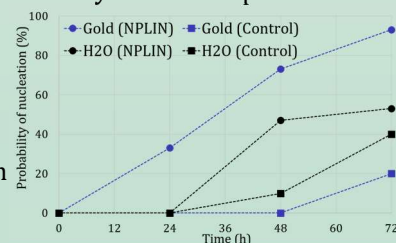
Nanoparticle heating seems the best hypothesis since presence or absence of particles has a strong influence on NPLIN behavior but it is not clear how particles are involved in the nucleation of the crystal (Hypothesis b, c or d from(4)).

Perspectives

Possibility to enhance the NPLIN effect by addition of particles.

- For Diprophylline/ 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.