Role of Solid Impurities on the Non-Photochemical Laser Induced Nucleation of Ethylenediamine Sulfate solutions

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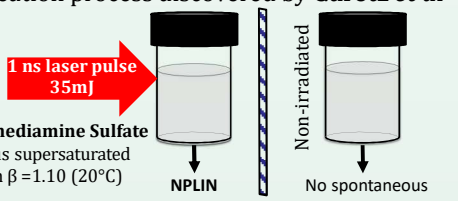
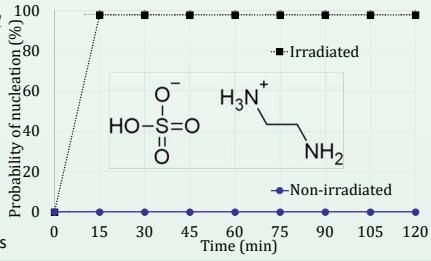
NPLIN = Non-Photochemical Laser Induced Nucleation

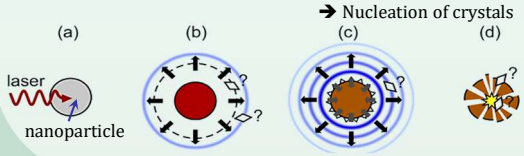
Nucleation process discovered by **Garetz *et al*[1]**





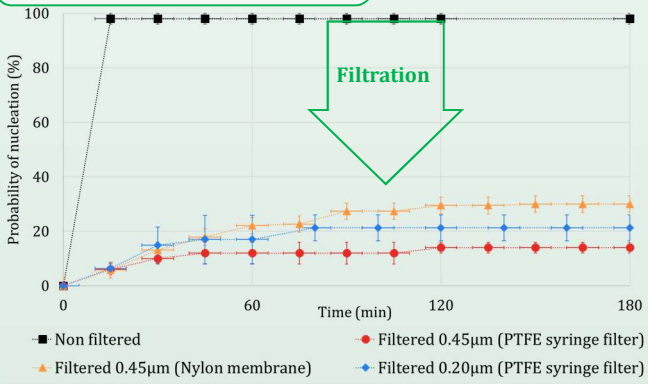
How does it work ?
Nanoparticle heating mechanism hypothesis

a) Absorption of laser light
b) Cavitation (vapor bubble)
c) Pressure waves
d) Destruction of the particle
➔ Nucleation of crystals



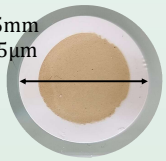
Adapted from (2)

Influence of Filtration



➔Evidence of the **presence of impurities** in the initial solution : Impurities responsible for the NPLIN phenomenon

➔Analysis of the filter and characterization of the impurities
➔Orange solid impurities recovered from filtration
➔ **ICP-OES** analysis results



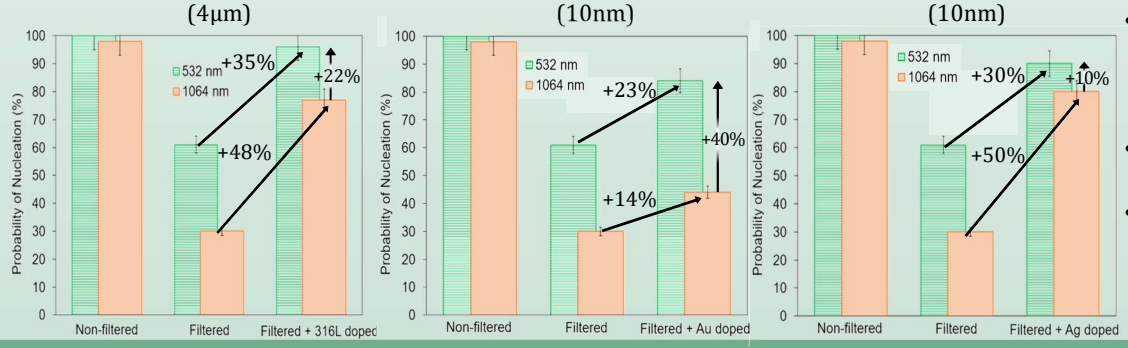
Concentration (mg/L)	Fe	Ni	Cr	S
Control	3.30	0.28	0.06	0.14
Impurity Sample	8.20	0.76	0.71	1.60

Metallic impurities ➔ Possibly **Stainless steel**

Influence of Doping and Wavelength

Doping of EDS filtered solution (β=1.10) with small metallic particles, results at 200min after irradiation(1 ns pulse, 35mJ, wavelength : 1064 nm or 532 nm)

316L **Stainless steel** powder (4µm) | **Gold** Nanoparticles (10nm) | **Silver** Nanoparticles (10nm)

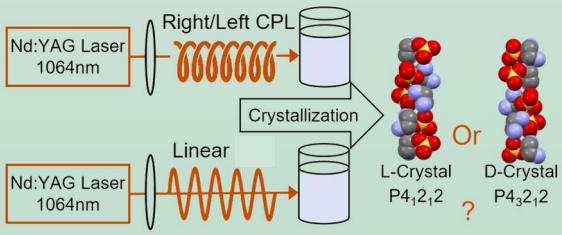


- **Solid metallic particles** in solutions required to retrieve high probability of nucleation with NPLIN
- Different behavior according to the nature of the doping particle
- **532nm** wavelength more efficient than **1064nm** (especially for gold that absorbs at 521nm)

Influence of Polarization geometry

Possible differences in particle absorption depending of the laser polarisation geometry

➔ Different NPLIN responses expected



Use of polarized light to control EDS crystal chirality

Experiment at β=1.10, 1pulse at 35mJ, 1064m

EDS solution	Polarization	N_{vial}	$N_{crystal}$	N_D	N_L	Cee±σ
Unfiltered	L-CPL	89	194	99	95	0.02±0.07
	LP	91	249	127	122	0.02±0.06
Filtered 316L-doped	L-CPL	12	20	9	11	-0.10±0.20
	LP	12	15	7	8	-0.10±0.30

L-CPL : Left Circularly Polarized light ; LP: Linear Polarized light

Similar results when using 532nm wavelength

➔ The change in laser polarisation shows **no significant change** in the Crystal Enantiomeric Excess (Cee) or nucleation probability.

Conclusions

- Filtration decreases NPLIN efficiency
- Doping increases NPLIN efficiency

} In favor of the Nanoparticle heating mechanism

- NPLIN efficiency impacted by the wavelength of irradiation
- No influence of laser polarization geometry on NPLIN

Perspectives

•Understand how particles are involved in the nucleation of the crystal to understand NPLIN mechanism
•Use of tailor made particle to trigger NPLIN in systems that are difficult to nucleate

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References
(1) Garetz, B. A.; Aber, J. E.; Goddard, N. L.; Young, R. G.; Myerson, A. S. *Phys. Rev. Lett.* **1996**, *77* (16), 3475 ;
(2) Alexander, A. J.; Camp, P. J. *J. Chem. Phys.* **2019**, *150* (4), 040901. ; (3) Briard, M.; Brandel, C.; Dupray, V. *Cryst. Growth Des* **2023**.