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Study of the dependence of the Amplified Spontaneous Emission (ASE) and the sensing properties on the capping ligand in $CsPbBr_3$ nanocrystals thin films

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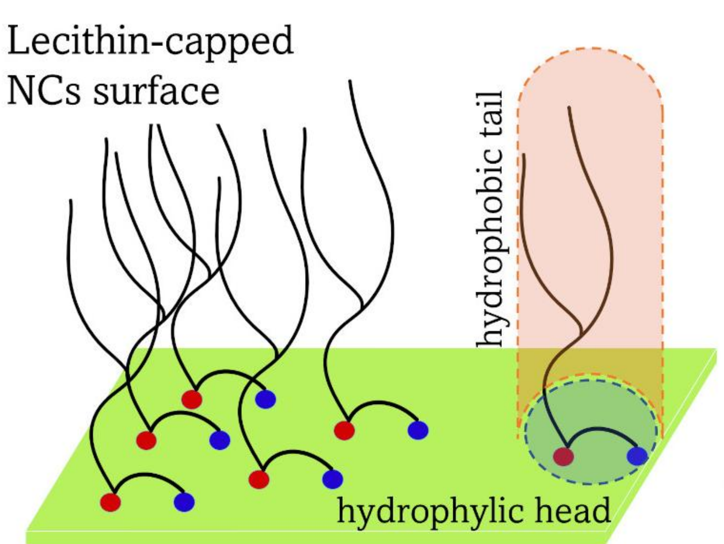
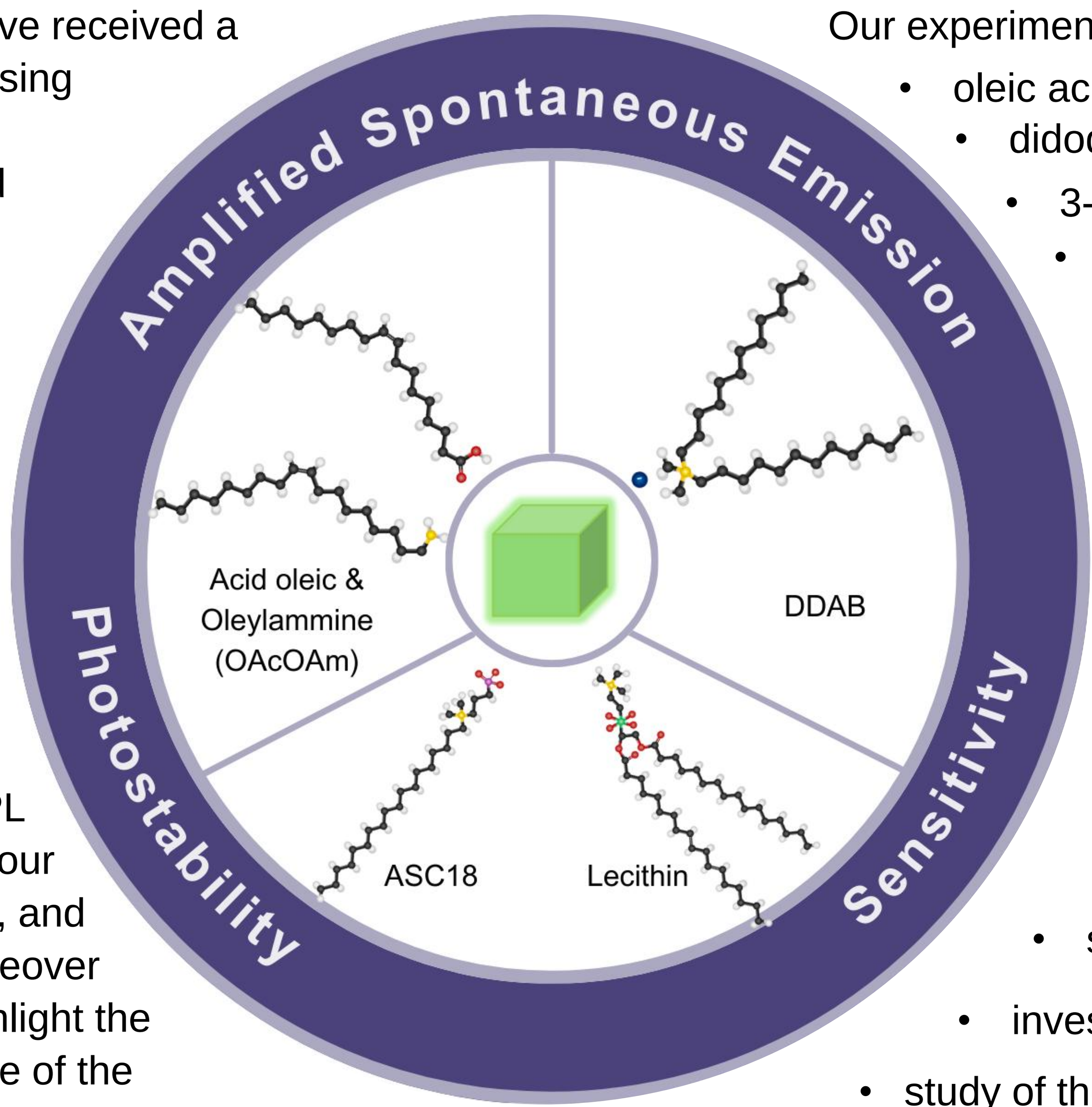
INTRODUCTION & AIM

Fully inorganic lead halide perovskite nanocrystals (NCs) have received a lot of attention as active materials for optoelectronic and sensing devices thanks to reversible environmental effects [1]. It has recently been demonstrated the sensitivity of lecithin-capped $CsPbBr_3$ NCs thin films to ambient air, noticeable as reversible modulation of the PL and ASE intensities, that rules out degradation effects. Moreover, air sensitivity of ASE was demonstrated to be up to 6.5 times higher than the spontaneous emission one [2]. However, the effect of the capping ligands on the ASE environmental sensitivity is to date fully unexplored.



Here we present a systematic study of the effects of the NCs capping ligand on the PL and ASE properties of four $CsPbBr_3$ NCs thin films, and their photostability; moreover for the first time we highlight the importance of the choice of the surfactant on the sensitivity to ambient air.



METHOD

Our experiments have been performed on four different samples

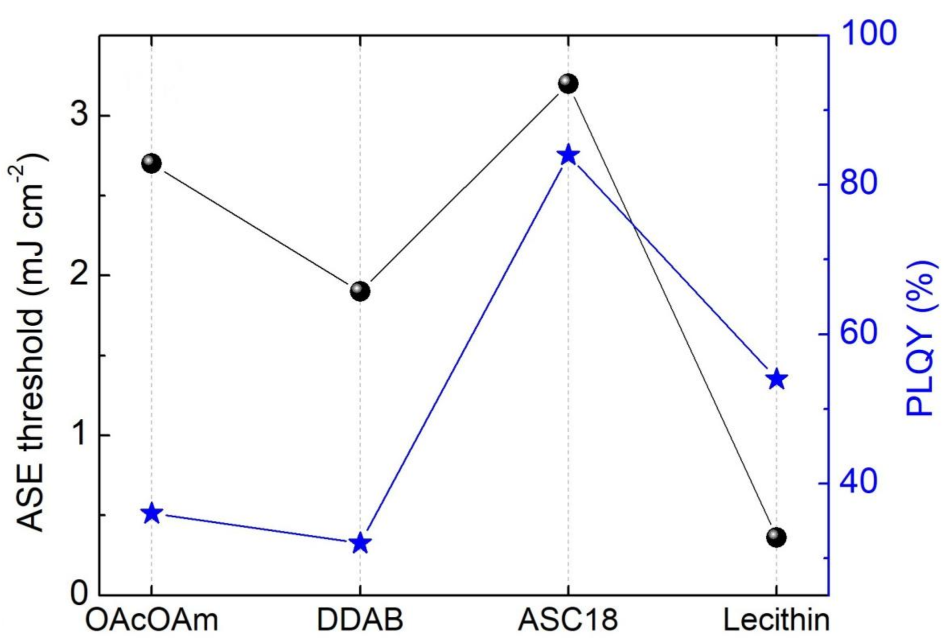
- oleic acid and oleylamine (OAc/OAm),
- didodecyldimethylammonium-bromide (DDAB),
- 3-(N,N-dimethyloctadecylammonio)propanesulfonate (ASC18)
- lecithin

Nanocrystals have been synthesized according to the recently developed approach described in ref [3]. Thin films have been deposited by spin coating NCs solution on quartz substrates at room temperature under chemical hood. Our experiments include:

- determination of the PLQY
- calculation of the ASE threshold
- determination of the optical losses coefficient
- study of the film morphology with SEM analysis
- investigation of the sensitivity to ambient air
- study of the photostability in air, under strong laser irradiation.

RESULTS & DISCUSSION

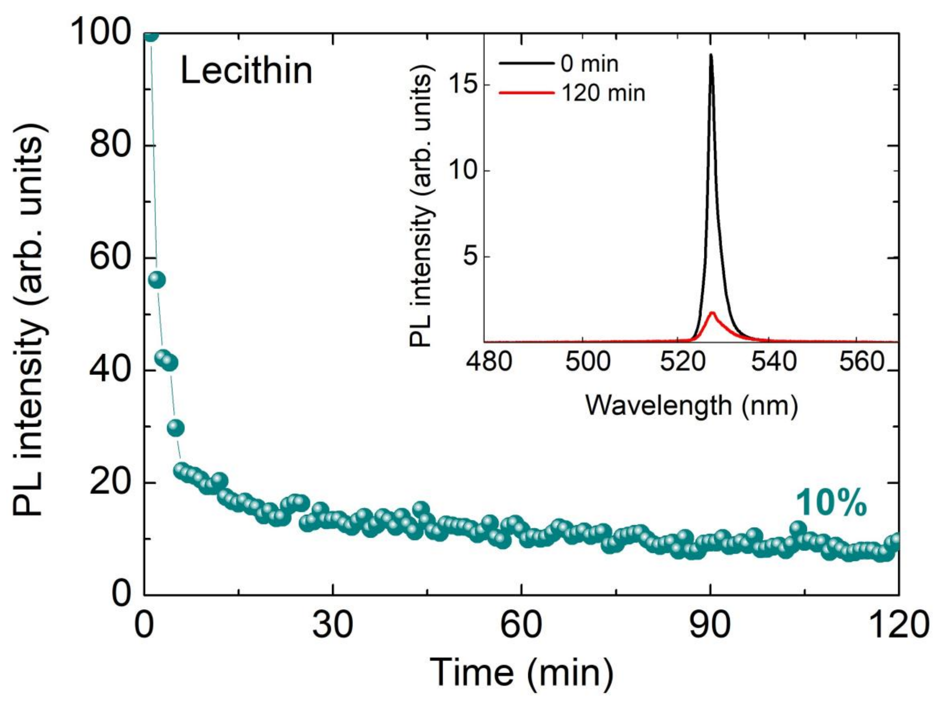
PL and ASE properties



- ASC18 and lecithin shows high PLQY thanks to good surface passivation
- OAcOAm and DDAB show low PLQY due to the weak ligand-perovskite bond that induces an easy ligand detachment

Room temperature ASE under ns excitation. The lowest threshold is detected for the lecitihin capped NCs film (0.36 mJ/cm^2), thanks to the lower optical losses and smoother film surface compared to the other samples.

Photostability

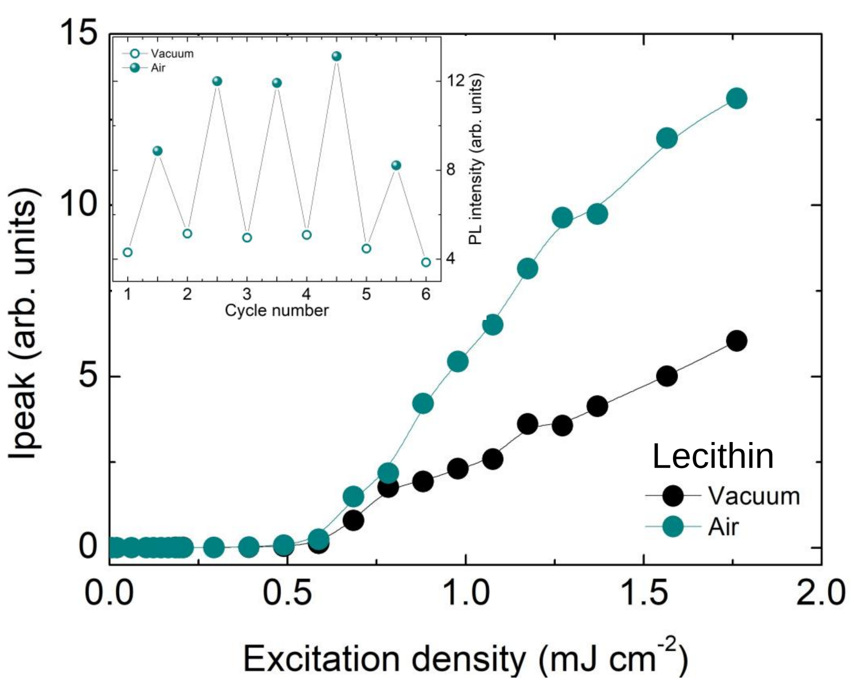


Continuous laser irradiation for 2h in air (about 1.5 times the ASE threshold).

- OAcOAm: it ends the kinetic with 60% of the initial ASE intensity
- DDAB and lecithin: show an initial rapid quenching followed by a slower ASE intensity decrease
- ASC18: ASE disappears after 80 minutes

Differences in the photostability are ascribed to **thermal conductivity** of the samples. A higher degree of surface ligand coverage prevents an efficient heat dissipation, inducing a temperature increase in the active material and a consequent rapid ASE quenching.

Air sensitivity



- OAcOAm: no sensitivity
- DDAB and ASC18: comparable sensing (20% RQ)
- Lecithin: strongest sensitivity (60% RQ)

Reversible perovskite-air interaction

The polar nature of the surfactants makes samples sensitive to the presence of water in the atmosphere. A facile deprotonation of the oleic acid in presence of OAm results in the formation of an acid-base complex, making the molecules overall neutral.

CONCLUSION

Summary of the analyzed properties (PLQY, ASE threshold and Sensitivity) as a function of the NCs capping ligand used. A color level has been assigned to each tab, from worst (red) to best (green). The choice of the ligand is of critical importance for the optical properties of the resulting film. $CsPbBr_3$ NCs thin films can represent valid candidates as active materials for ASE-based sensing devices as long as a proper design of the nanocrystals surface passivation is actuated.

	OAcOAm	DDAB	ASC18	Lecithin
PLQY	36%	32%	84%	54%
ASE threshold	2.7 mJ/cm^2	1.7 mJ/cm^2	3.2 mJ/cm^2	0.36 mJ/cm^2
Sensitivity	0%	20%	20%	60%

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