Supporting Materials (SM)

Polarized luminescence in single upconversion $NaYbF_4$: Er rod

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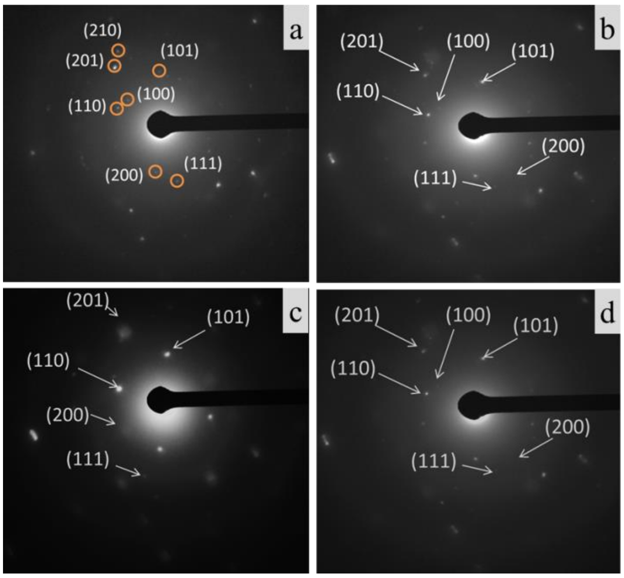

Fig. S1. Selected area electron diffraction (SAED) patterns captured from hexagonal $NaYbF_4$:Er(2%) rods deposited on a silicon substrate.

In Fig. S1 (a-d), the diffraction patterns are characteristic for hexagonal phase $NaYbF_4$ nanocrystals. Strong and defined reflections corresponding to crystal planes (100), (110), (101), (111), (200) and (201) are indexed, positively indicating pure hexagonal phase $NaYbF_4$.

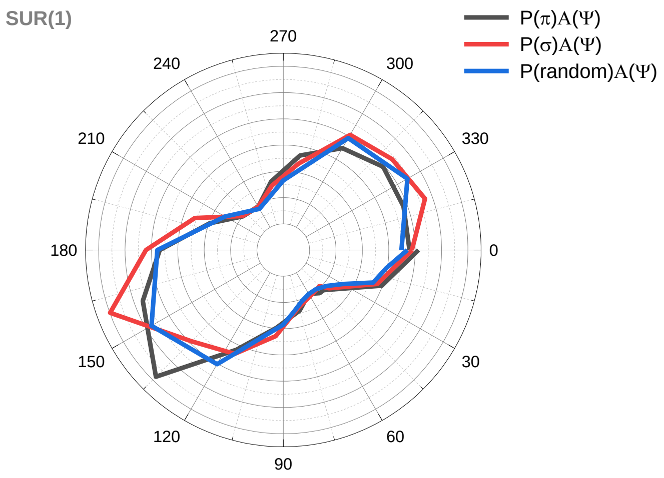

Fig. S2. Analyzer angle (Ψ) dependence of the polarization response defined as $R = I_{655\,nm}/I_{666\,nm}$ at the parallel P(π) (black) and perpendicular P(σ) (red) laser excitation polarization toward SUR(1) c-axis (P(π)A(Ψ) and P(σ)A(Ψ) configurations, respectively). Ψ is the analyzer angle in the COM coordinate system.

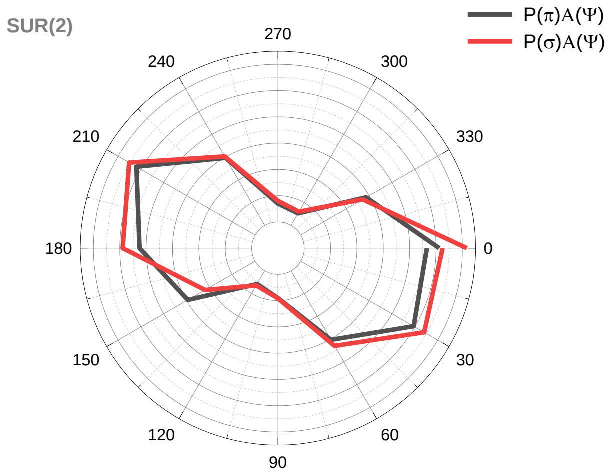

Fig. S3. Analyzer angle (Ψ) dependence of the polarization response defined as $R = I_{655\,nm}/I_{666\,nm}$ at the parallel P(π) (black) and perpendicular P(σ) (red) laser excitation polarization toward SUR(2) c-axis (P(π)A(Ψ) and P(σ)A(Ψ) configurations, respectively). Ψ is the analyzer angle in the COM coordinate system.

Modelling polarization responses of SUR and a small SUR ensemble

In order to explain basic difference between polarization responses of SUR and a small ensemble of isotropically oriented rods, let us introduce a simple model, when the spectrum of SUR consists of two Gaussian bands only, centered at 655 nm and 666 nm:

$$G_{\lambda_0}(\lambda) = exp\left(\frac{(\lambda-\lambda_0)^2}{2\cdot\delta\lambda^2}\right).$$

According to our experimental findings, the SUR emission exhibits pronounced polarization anisotropy. Then, let us assume, that the 655 nm band is π-polarized, and 666 nm band is σ-polarized. It allows constructing the following fitting function with angle-dependent spectral intensities:

$$S(\lambda,\theta) = (a + cos(\theta)^2)G_{655}(\lambda) + (b + sin(\theta)^2)G_{666}(\lambda),$$

where $a = 1.8$ and $b = 1$ are fitting parameters. Varying these constants we strive for the closest as possible resemblance of the modeled spectrum profile (Fig.S4) to the experimental data in Fig 5 (a).

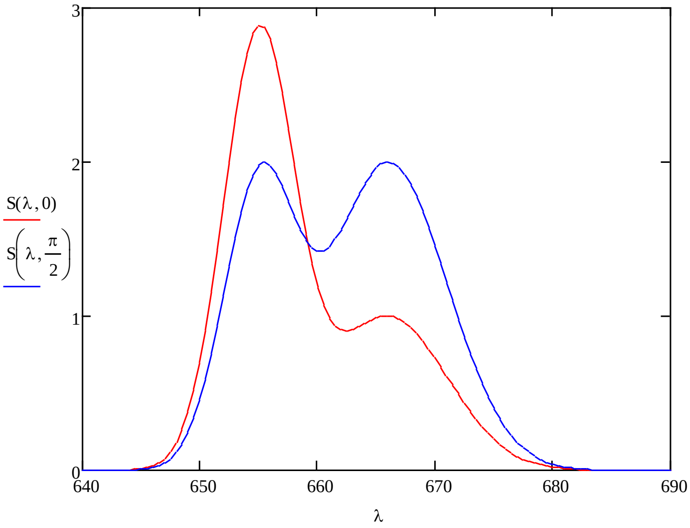

Fig. S4. The fitting function $S(\lambda,\theta)$ at angles $\theta = 0$ (red line) and $\theta = \frac{\pi}{2}$ (blue line), which correspond to π- and σ-polarizations of the SUR emission.

One may see (Fig.S5), that the polarization response defined as $R(\theta) = \frac{S(655,\theta)}{S(666,\theta)}$ demonstrates the same angular tendency as the measured polarization response in Fig 7.

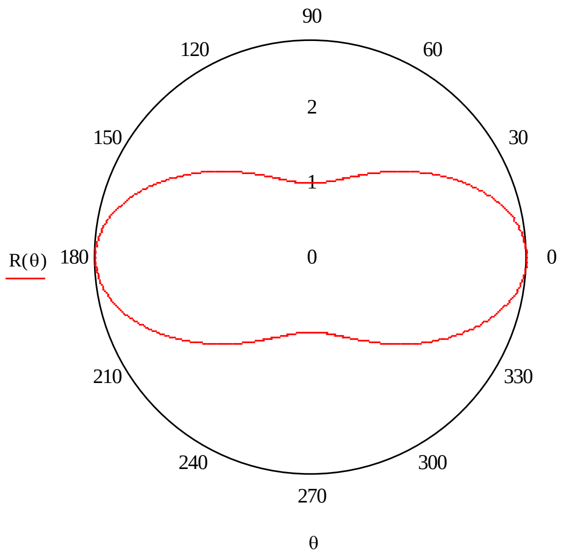

Fig. S5. Calculated *R*-polarization response for SUR.

The group of randomly-oriented SURs presented in Fig 9 could be roughly considered as equilateral triangle, to which a following fitting function may be attributed:

$$S3(\lambda,\theta) = S(\lambda,\theta) + S\left(\lambda,\theta + \frac{2}{3}\pi\right) + S\left(\lambda,\theta + \frac{4}{3}\pi\right).$$

In this case $S3(\lambda,\theta)$ remains constant for any angle θ (Fig.S6), hence the polarization response $R3(\theta)$ is completely isotropic (Fig.S7).

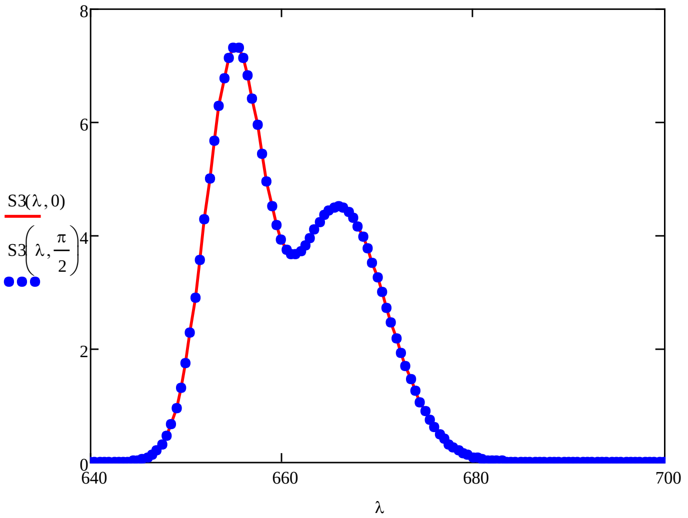

Fig. S6. The fitting function $S3(\lambda,\theta)$ at angles $\theta = 0$ (red line) and $\theta = \frac{\pi}{2}$ (blue dots), which corresponds to the emission of an ensemble of three isotropically oriented SURs.

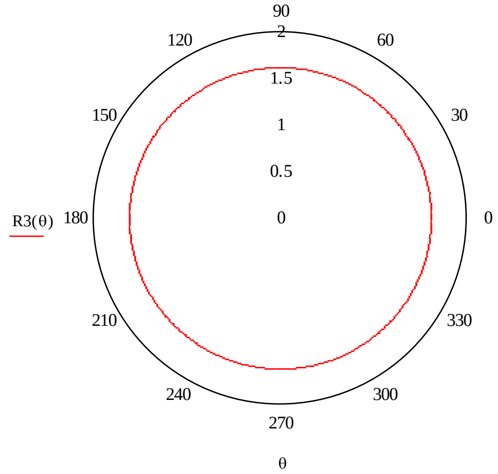

Fig. S7. Calculated *R3*-polarization response of three isotropically oriented SURs.

This simple model does not describe all the features typical for the emission anisotropy of real SURs, however it clarifies why the luminescent response of a small ensemble of isotropically-oriented particles could cease to be polarized.

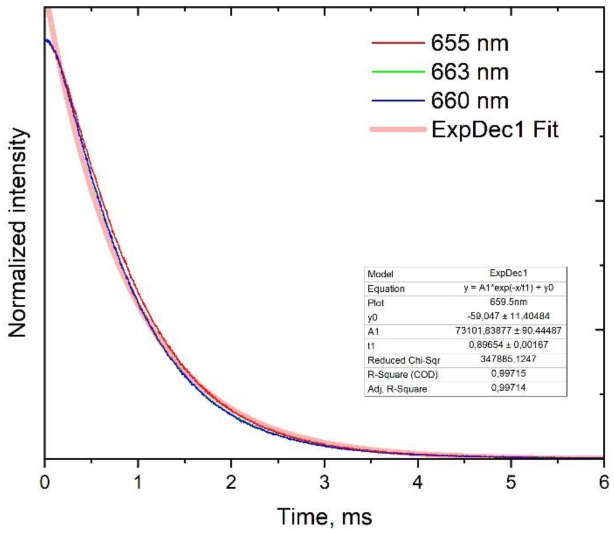

Fig. S8. Luminescence kinetics measured at room temperature with laser excitation at 980 nm. Performed fitting shows that decay character of luminescence spectral components at 655, 660 and 663 nm is close to mono-exponential with decay time of 896 μs.