Supporting Information

Single-particle Polarized Spectroscopy Determines Upconverting Nanorod Orientation During 3D Optical Manipulation.

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S1.- Dynamic light scattering of the studied sample

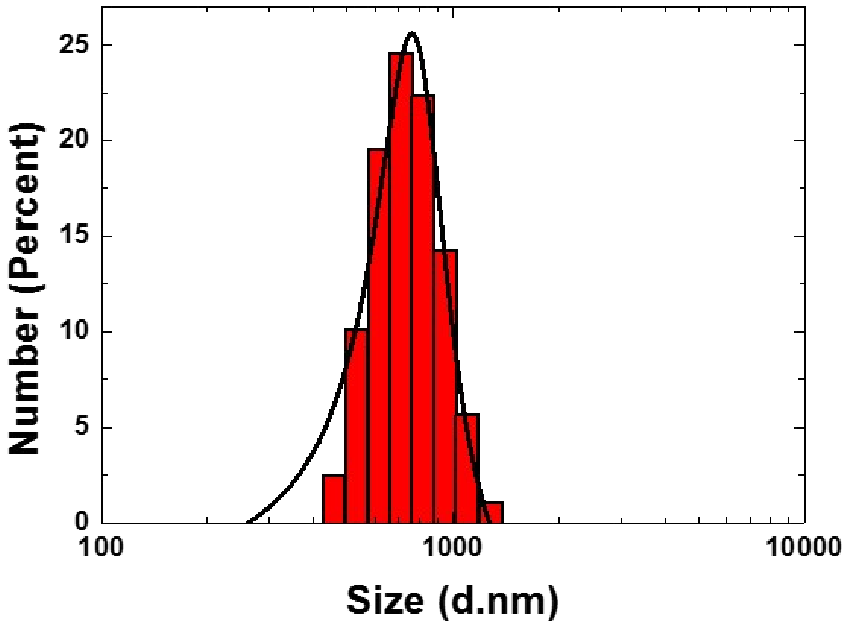

Fig. S1. Dynamic light scattering diagram.

We have performed the DLS measurements for the $NaYF_4:Er^{3+},Yb^{3+}$ UCNRs dispersed in water in optically homogeneous square polystyrene cell using a Zetasizer Nano from Malvern Instruments with the detection angle of 173°. The measurements were performed at 25°C, and the hydrodynamic diameter (D_H) and polydispersity index (PdI) values were obtained as an average of three runs with at least 10 measurements. The DTS (Nano) program was applied for data evaluation. It must be kept in mind that the analysis of DLS data assumes the spherical shape of NPs, and thus the obtained D_H value for studied NRs can be only treated as a rough estimation. Nevertheless, the DLS measurements showed the mean size of NRs of around 1µm (see Fig S1), and low PdI value of 0.290, which confirms the colloidal stability and absence of aggregates in the solution.

S2.- Optical manipulation by two beam optical tweezers setup

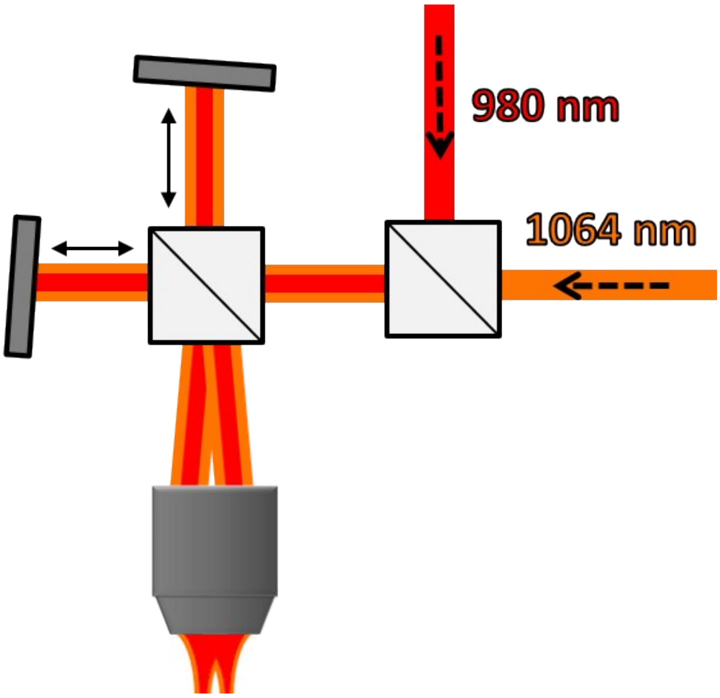

Fig. S2. Modification of the optical tweezers setup for two-beam optical trapping experiments.

S3.- Polarized emission of the colloidal suspension of $NaYF_4$:Er^{3+},Yb^{3+} UCNRs.

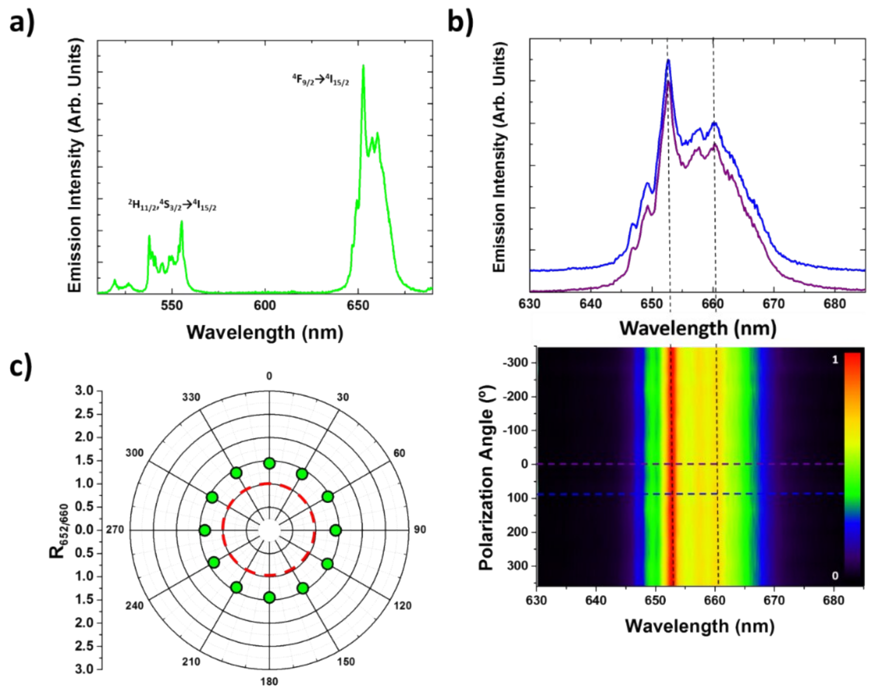

Fig. S3. (a) Emission spectrum of green and red emission bands for the colloidal suspension of $NaYF_4$:Er^{3+},Yb^{3+} UCNRs (10^{11} NPs/cm^3). **(b)** Emission spectra of the red band for two perpendicular emission polarization angles for the colloidal suspension of $NaYF_4$:Er^{3+},Yb^{3+} UCNRs. Emission is a superposition of the parallel and perpendicular to the UCNR optical axis emissions. In the lower part, it is also shown the evolution of the normalized emission intensity with emission polarization angle. **(c)** Polar plot of the ratio between 652 and 660 peak intensities as a function of the emission polarization angle. Red dashed circle delimits under and above 1 ratio values.

S4.- Green emission band.

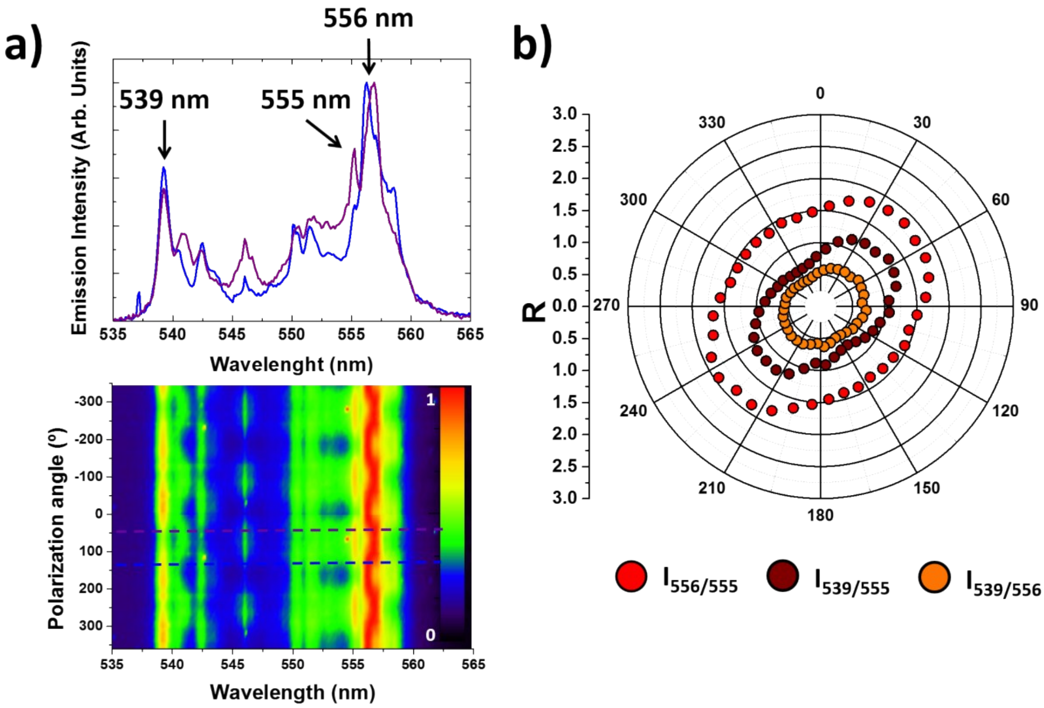

Fig. S4. (a) Emission spectra from a single $NaYF_4$:Er^{3+},Yb^{3+} UCNR immobilized on a surface for two perpendicular emission polarization angles. In the lower part, evolution of the emission intensity with emission polarization angle is also shown. **(b)** Polar plot of the ratio between peak intensities as a function of the emission polarization angle.

S5.- Force measurements.

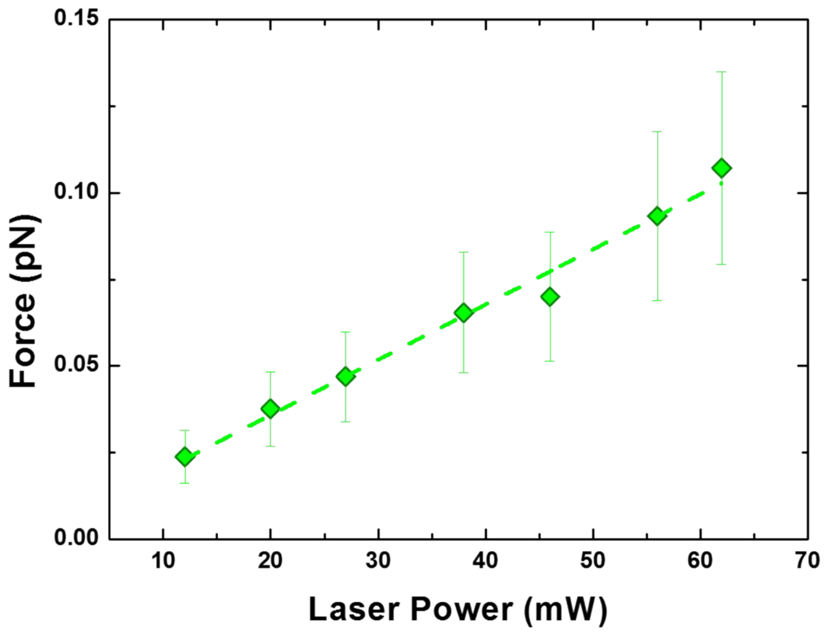

Fig. S5. Optical force as a function of the laser power calculated by the hydrodynamic drag method. Green diamonds represent the friction force over a cylinder of 140 nm in diameter and 1100 nm long calculated as in Gao and coworkers work. [1]

The hydrodynamic drag method was used to measure the optical forces exerted over the $NaYF_4$:Er^{3+},Yb^{3+} UCNRs. It is based on the drag force that experiments an object, i.e. a particle, moving through a fluid. This drag force is directly proportional to the relative velocity between the particle and the fluid and it opposes its movement. For low velocities, the optical trapping force is larger than the drag force and the particle remains trapped. In this regime, the optical force can be considered as a restoring force ($F_{trap} = \kappa_{trap} r$) which tends to return the particle to the equilibrium position inside the optical trap. The strength of the optical trap, κ_{trap}, depends on microscope objective NA (0.80) and laser power. When the velocity over takes the maximum velocity, v_{max}, the drag force becomes larger than the optical trapping force so that the particle escapes from the trap. The largest distance from the trap center at which the particle is still trapped corresponds to the trap radius. This point is reached when optical trapping and drag force equalizes at the maximum velocity. This fact allows for obtaining a value for the strength of the optical trap by moving the trapped particle in a controlled way. This experiment was performed by displacing the microchannel, where rod colloidal suspension was placed, along its longitudinal axis by using a motorized stage. This produces a relative velocity between the trapped particle and the fluid allowing for finding the maximum velocity at which the particle is still trapped.

An accurate expression for the drag force over a cylinder moving throw a fluid is given in Gao and coworkers work.[1] In Fig S5 a representation of the as calculated force is plotted as a function of the laser power. Optical trap strength is calculated to be $(2.167 \pm 0.002) pN/W\mu m$.

References:

1. W. Gao, S. Sattayasamitsathit, J. Orozco and J. Wang, *Journal of the American Chemical Society*, 2011, **133**, 11862-11864.