Supporting Information

Sub-millisecond electric field sensing with an individual rare-earth doped ferroelectric nanocrystal

Athulya K. Muraleedharan,[†] Jingye Zou,[‡] Maxime Vallet,[‡] Abdelali Zaki,[‡]

Christine Bogicevic,[‡] Charles Paillard,^{‡,¶} Karen Perronet,[†] and François

Treussart*^{,†}

† Université Paris-Saclay, ENS Paris-Saclay, CNRS, CentraleSupélec, LuMIn, 91190 Gif-sur-Yvette, France

‡Université Paris-Saclay, CentraleSupélec, CNRS, Laboratoire SPMS, 91190 Gif-sur-Yvette, France

¶Smart Ferroic Materials, Institute for Nanoscience & Engineering and Department of Physics, University of Arkansas, Fayetteville 72701 Arkansas, USA

E-mail: francois.treussart@ens-paris-saclay.fr



Figure S1: Size and shape distributions of nanoBTO:Yb,Er. (a) Particle size distribution as inferred from SEM images (size defined as the arithmetic mean of the side lengths of the rectangular shaped nanocrystals) with an average value of 158 ± 27 nm. (b) Aspect ratio, revealing a majority of particles with a ratio of 1 (cubic shape). Inset: SEM images (30 kV acceleration voltage and $100000 \times$ magnification) of single NCs with aspect ratio 1 and 0.75; scale bars: 100 nm.



Figure S2: Rietvelt refinement analysis on nanoBTO:Yb,Er. Experimental diffractogram data (red open circles), modeled diffractogram (black line), small green vertical bars below the diffractogram indicate $hk\ell$ positions and the blue curve beneath represents the difference between the data and the model. The refinement was carried out using the Win-Plotr/FullProf package.¹ The peak shape was described by a pseudo-Voigt function, and the background level was modeled using a polynomial function.



Figure S3: Variation of intensity of the green and red up-conversion bands with the laser excitation power. Total area of all the green transition bands (green squares, $(I_{\rm UC})^{\rm Green}$) and the red transition band (red circles, $(I_{\rm UC})^{\rm Red}$) as a function of the infrared laser (977 nm wavelength) excitation laser P. Lines are fits to a power law. $(I_{\rm UC})_{\rm red}$ has a linear dependence with P.

Table S 1: Different responses to positive or negative applied voltage for the eight nanoBTO:Yb,Er (same numbering as in the main text) displaying fast upconversion intensity variation. The checkmark indicates observed fast variation (within one time bin duration of 100 μ s), while "slow" means a response lasting up to a few seconds.

	1	2	3	4	5	6	7	8
UC \nearrow with +10 V	\checkmark		\checkmark					
UC \searrow with +10 V				slow	\checkmark	slow	\checkmark	\checkmark
UC \nearrow with -10 V		\checkmark		\checkmark	slow	\checkmark	\checkmark	
UC \searrow with -10 V								



Figure S4: Absence of UC spectrum modification on nanoBTO:Yb,Er due to the AFM tip mechanical force applied at the contact. Spectra recorded at 10 mW excitation laser power and 10 s exposure duration. We do not see any difference between the NC spectrum acquired with the tip not in contact (dark grey) and with the tip in contact with no applied voltage (red).



Figure S5: Absence of UC spectrum modification under applied voltage in nonferroelectric Y_2O_3 :Er nanocrystals. (a) AFM topography image of a small aggregate. The particle surrounded by the dashed circle is the one on top of which the conductive tip is placed to apply voltage. (b) Up-conversion spectra of aggregate shown in (a), at 400 μ W excitation laser (977 nm wavelength) power and with 1 s exposure duration, in various conditions: no applied voltage (black), +10 V applied (red) and -10 V (blue). (c) Total UC intensity time trace during the square variation of applied voltage between 0 and 10 V displayed on top.



Figure S6: Absence of UC spectrum modification under applied voltage in nonferroelectric NaYF₄:Yb,Er nanocrystals. (a) AFM topography image of a small aggregate of 500×65 nm NaYF₄:Yb,Er nanorods. (b) Up-conversion rasterscan of the same aggregate as in (a) at 77 μ W excitation laser (977 nm wavelength) power. (c) UC spectrum at the same excitation laser power as in (b), and 10 s exposure duration, with or without applied 10 V voltage.



Figure S7: Examples of other up-conversion signal variation behaviors with applied electric field, differing from the fast variation with constant SHG. (a) Example of a nanocrystal with no variation of UC or SHG (inset) intensities whatever the polarity of applied voltage is. This NC has a height of 133 nm. (b) Example of a nanocrystal for which the UC intensity varies (decreases) with a slow response time when -10 V is applied, and without any change in SHG signal. (b1): UC and SHG peak (inset) spectra. (b2): total UC intensity time trace function of applied voltage. (c) Example of a nanocrystal with fast variation of UC intensity time trace function of applied voltage. (d) Example of a nanocrystal with fast slow variation of UC intensity and SHG signal. Left (d1): UC and SHG peak (inset) spectra. Right (d2): total UC intensity time trace function of applied voltage.

References

 Rodríguez-Carvajal, J. Recent advances in magnetic structure determination by neutron powder diffraction. *Physica B: Physics of Condensed Matter* 1993, 192, 55–69.