Electronic Supplementary Material (ESI) for Soft Matter.

Fibrillogenesis from Nanosurfaces: Multiphoton Imaging and Stereological Analysis of Collagen 3D Self-assembly Dynamics

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Supplementary Information

2PEF signals from other fluorophores and particles

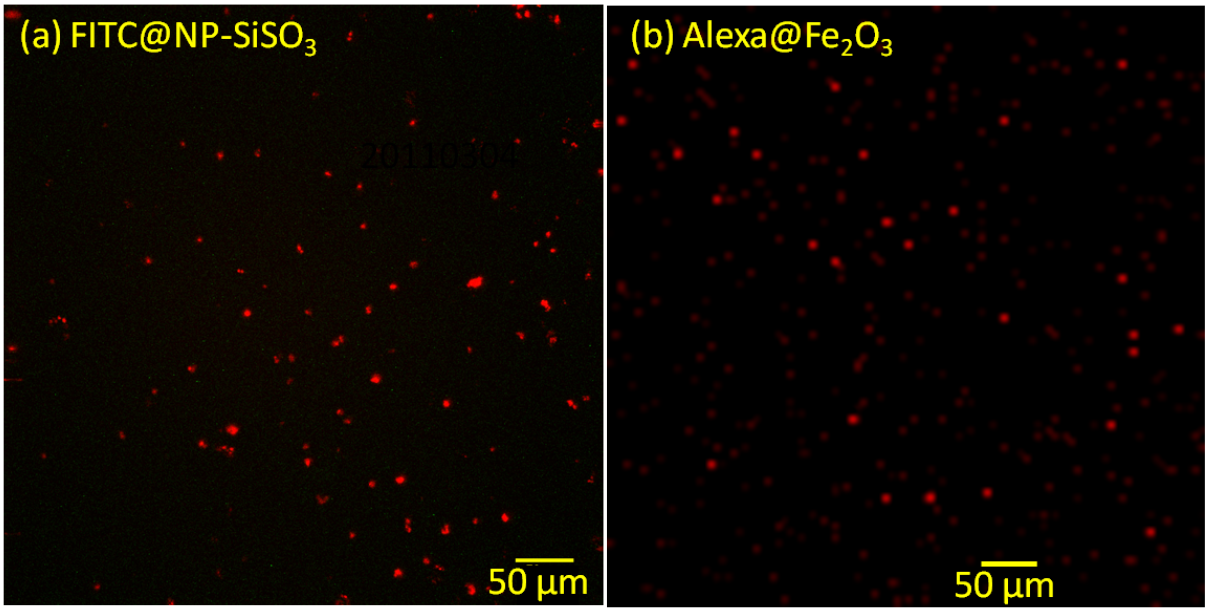

Fig. S1. 2PEF signals from other fluorophores and particles. (*ca*. 300 nm diameter) (a) Silica particles functionalized with fluorescein isothiocyanate (FITC), and (b) iron oxide particles loaded with Alexa Fluor® 488. This illustrates the fact that 2PEF should be widely applicable to any kind of particles and fluorophores.

SHG photon density around particles.

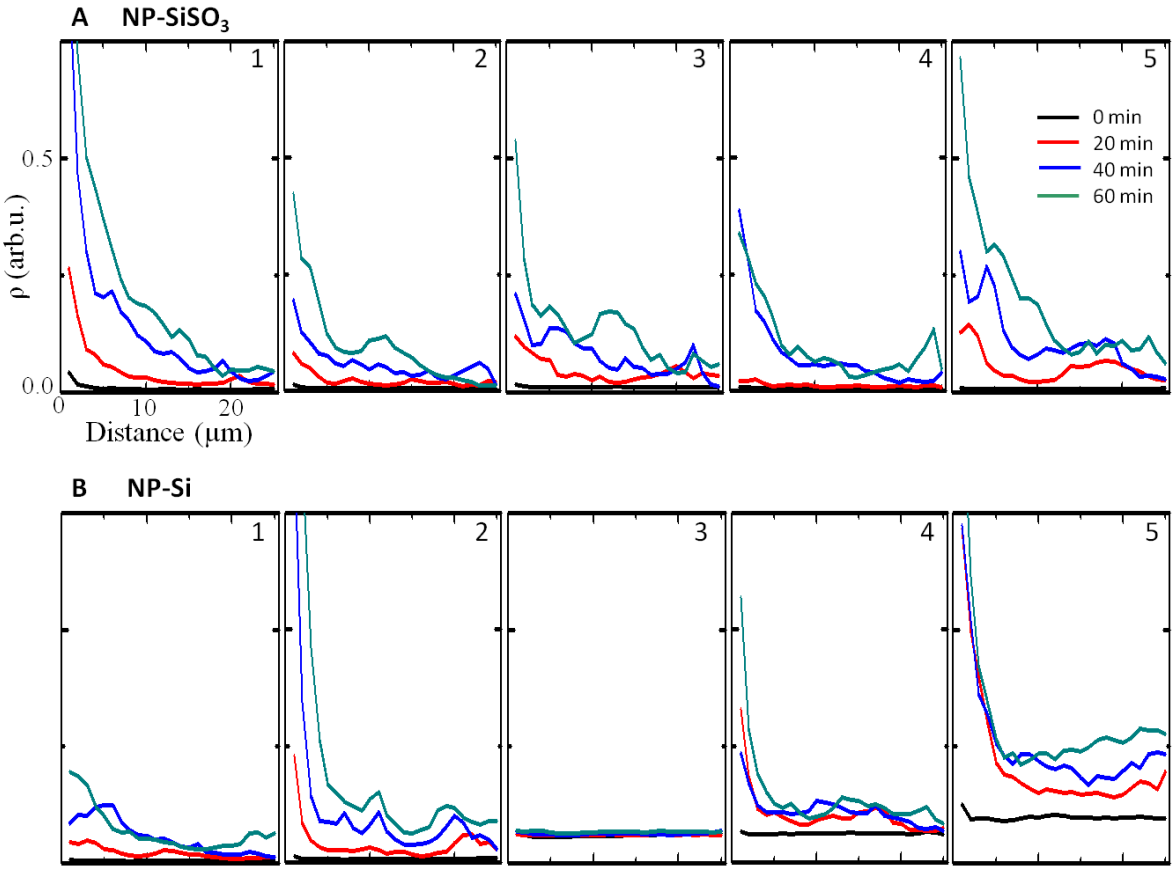

Fig. S2. SHG photon density around particles. The computation of $\rho(C_j)$ has been done at 4 sequential times, for 5 samples involving NP-$SiSO_3$ particles (Fig.S1 *A1-5*) and 5 NP-Si samples (Fig.S1 *B1-5*). One can observe that SHG photon density increases with time. The evolution is highly reproducible in the case of functionalized NP-$SiSO_3$, where the SHG photon density is always maximal close to the particle surface and exponentially decays with increasing distance. In contrast, NP-Si density curves exhibit fluctuating behaviours, sometimes without any maximum in SHG photon density or with non-monotonic variation with respect to the particle surface. This indicates that there is no systematic correlation between the presence of non-functionalized NP-Si surfaces and the course of fibrillogenesis.

Quantitation of collagen fibrillogenesis using a statistical approach.

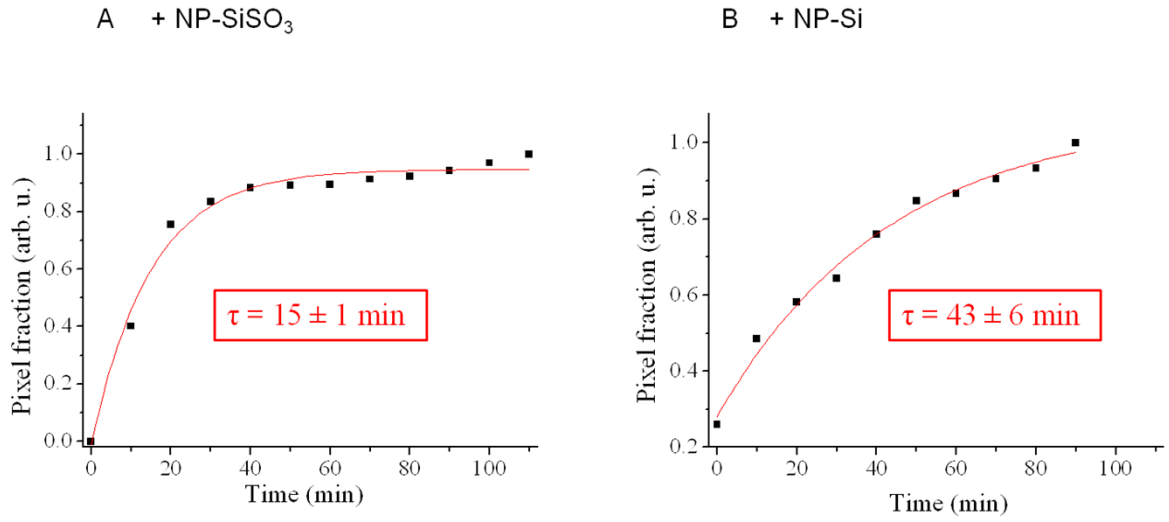

Fig. S3. Quantitation of collagen fibrillogenesis using a statistical approach. Fibril volume density (pixel fraction) over time for collagen fibrillogenesis in presence of (A) functionalized NP-$SiSO_3$ (τ = 13 ± 3 min over three samples) and (B) bare NP-Si (τ = 30 ± 8 min over three samples). Black squares and red lines correspond to experimental data and exponential fitting respectively.

TEM photos of collagen/NP-$SiSO_3$ samples

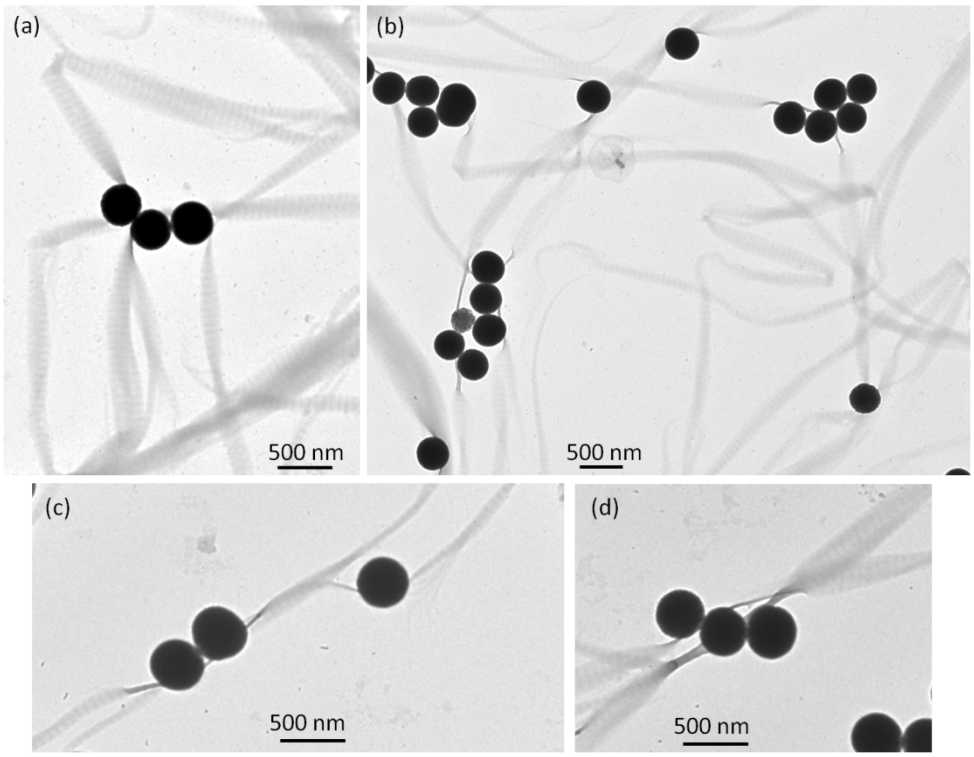

Fig. S4. TEM photos of collagen/NP-$SiSO_3$ samples. This set of photos shows (a-b) the presence of a discrete number of collagen fibrils with the expected banded pattern, their extent to a micron scale, and (c-d) their interaction with the particle surface observed as a dense rod of protein on the nascent collagen fibril.