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Electronic Supplementary Information



Fig. S1 (a) Plane-view high-angle annular dark-field scanning transmission electron microscopy image of a nAAA-5 trilayer. The areal density of Ag NPs is of the order of $2000 \,\mu\text{m}^{-2}$ and the period in the transverse direction (\perp) is $\Lambda_{\perp} = 36.3 \,\text{nm}$. The autocorrelation function taken from a zone of $1.2 \times 1.2 \,\mu\text{m}^2$ is shown in inset. (b) Distribution of interparticle gaps in the longitudinal direction (||); the median value is 5.1 nm. (c) In-plane size distributions in the transverse and (d) longitudinal directions. Corresponding average values \bar{x} and standard deviations σ_i are also given.



Fig. S2 Raman spectra excited at 514.53 nm of aqueous solutions of bipyridine: (black spectrum) a stock-solution of 8×10^{-3} M without enhancement effect and (red spectrum) a 10^{-8} M bipyridine dilution in contact with a colloidal dispersion of Ag NPs (15 nm average diameter).



Fig. S3 Raman spectra excited at 514.53 nm of colloidal suspensions of bipyridine (around 1×10^{-6} M) in contact with Ag NPs deposited on a silica slide: (black) excited with a low power (50 μ W μ m⁻²), the 240 cm⁻¹ band characterises the direct interaction between bipyridine molecules and Ag NPs, and (red) by multiplying the excitation power by 3, the spectrum evolves to reveal new signals, characteristic of amorphous carbon phase.



Fig. S4 SERS spectrum excited at 514.53 nm of a 10^{-5} M bipyridine dilution in contact with a colloidal dispersion of Ag NPs (15 nm average diameter) functionalized by a thiol-functionalized C₁₂ spacer to avoid any chemical bond between Ag-surface and bipyridine: absence of the 240 cm⁻¹ band.