Electronic Supplementary Information:

Experimental Section

Synthesis of solutions of HMBP coated nanocrystals

To prepare non-coated γ-Fe_2O_3 particles, the first step is to add dimethylamine ($(CH_3)_2NH$) to an aqueous micellar solution of ferrous dodecyl sulphate ($Fe(DS)_2$). The final concentrations after mixing of the reactants are 1.3×10^{-2} mol.L^{-1} and 8.5×10^{-1} mol.L^{-1} for $Fe(DS)_2$ and dimethylamine, respectively. The solution is stirred vigorously for 2 h at 28.5 ° C and the resulting precipitate of uncoated nanocrystals is isolated from the supernatant by centrifugation. In the second step, this precipitate is washed with an acidic solution (HCl 10^{-1} mol.L^{-1}) and a solution of HMBP molecules in water ([HMBP] = 1.4×10^{-1} mol.L^{-1}) is added. The solution is stirred for two hours at room temperature. The precipitate that appears is washed with an acidic solution (HCl 10^{-1} mol.L^{-1}) and the powder is dried in air. The magnetic nanocrystals coated with HMBP molecules are dispersed in water. The initial pH is equal to 2 and then progressively increased to pH 7 by addition of sodium hydroxide NaOH (10^{-1} mol.L^{-1}). The ferrofluid is dialyzed through a 10 000 g mol^{-1} membrane in order to exclude the uncomplexed HMBP molecules.

To test an eventual desorption of HMBP molecules, the pH of ferrofluids initially at pH=5; 7 and 10 is decreasing at a value of 3 by addition of an acidic solution (HCl). This induces nanocrystal precipitation. The supernatant is dried and after addition of D_2O, the solution is analyzed by NMR. No signal due to HMBP molecules is detected. This indicate no significant loss of surfactant molecules on nanocrystals surface, confirming previous considerations about the high binding of phosphonate and bisphophonate with iron[23, 24, 26].

Stability tests of HMBP coated nanocrystals

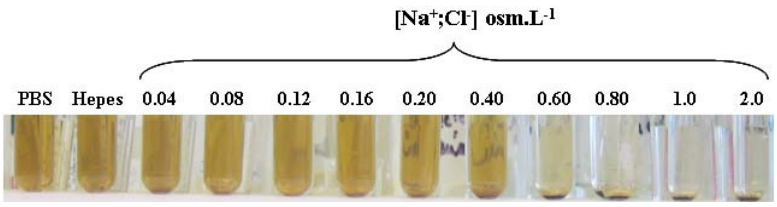

Figure S1. Behavior of the HMBP coated nanoparticles solution as function of salt concentration. The initial conditions are at pH 7.4 and iron concentration of 2 x 10^{-3} mol.L^{-1}. The salt concentration is adjusted with NaCl solution. Above 0.6 osm.L^{-1} , a macroscopic phase separation is observed through the apparition of a black precipitate (bottom phase). HMBPUSPIO are also stable within biological buffers (PBS and Hepes).

Figure S2. Behavior of the HMBP coated nanoparticles solution as function of pH. The initial conditions are at pH 7.1 and iron concentration of 4 x 10^{-3} mol.L^{-1}. %. The suspension pH is then adjusted with sodium hydroxide (NaOH) or acidic (HCl) solutions . Below pH 4.0, a macroscopic phase separation is observed through the apparition of a black precipitate (bottom phase).

HMBP coated nanocrystals as precursor group for functional molecules coupling

Standard procedures were used to crosslink free carboxylic acid group with amino containing ligands such as amino fluorescein. The HMBP coated nanocrystals were activated for 2 hours with 1-ethyl-3-(3-dimethyl aminopropyl) carbodimide (EDC) at pH 5 and a ratio [EDC]/[BP] equal to 10. Then, the activated nanocrystals were reacted with amino fluorescein for 2 hours at pH 7 and a ratio [dye]/[BP] equal to 3.

In order to exclude the uncomplexed dye molecules, the ferrofluid is dialyzed through a 10 000 g mol^{-1} membrane and then lyophilized. The powder is easily dispersed in water at pH7.

Chemical decomposition of the coupling between amino-fluorescein and the particle is realized mixing the solution at pH12 during 2h. The magnetic particles were then separated from the fluorescent supernatant by magnetic decantation.

The resulting washed products are characterized ATR-FTIR, FTIR, UV/Vis and Fluorescence spectroscopies. The samples for TEM were prepared by direct evaporation of the washed solution on an amorphous carbon TEM grid.

HMBPUSPIO calibration curve

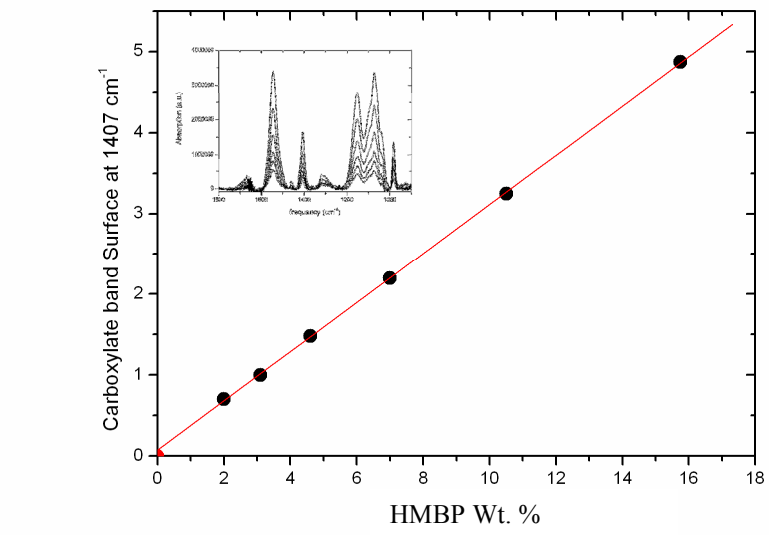

Figure S3. Calibrate curve of the carboxylate band surface at 1407 cm^{-1} as a function of HMBP weight concentration obtained from ATR-FTIR spectroscopy (insert).

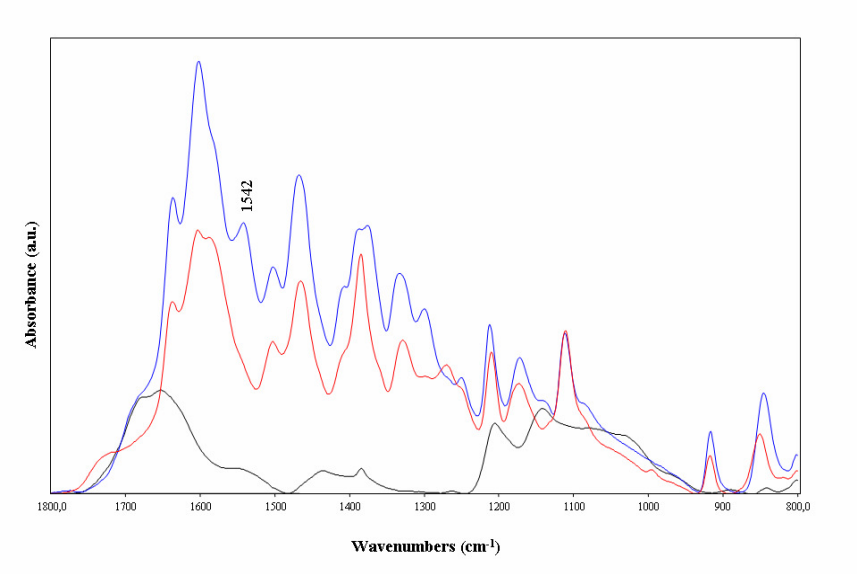

Figure S4. FTIR spectroscopy in KBr pellets: HMBP coated nanoparticles (black curve), pure amino-fluorescein (red curve) and amino-fluorescein coupled to HMBP coated nanoparticles (blue curve).

Citotoxicity tests

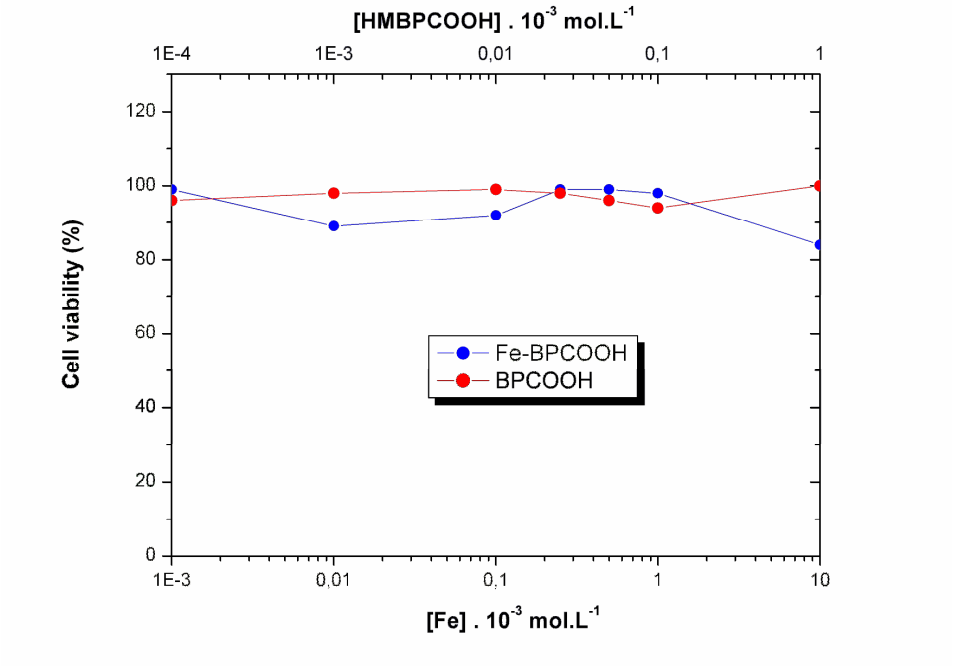

Figure S5. Viability tests of HMBP coated nanocrystals (blue curve) and free HMBP molecules (red curve) incubated with mammal cells (MDAMB-231) for 48 h.

HMBP coated nanocrystals incorporation into MDA-MB-231 breast cancer

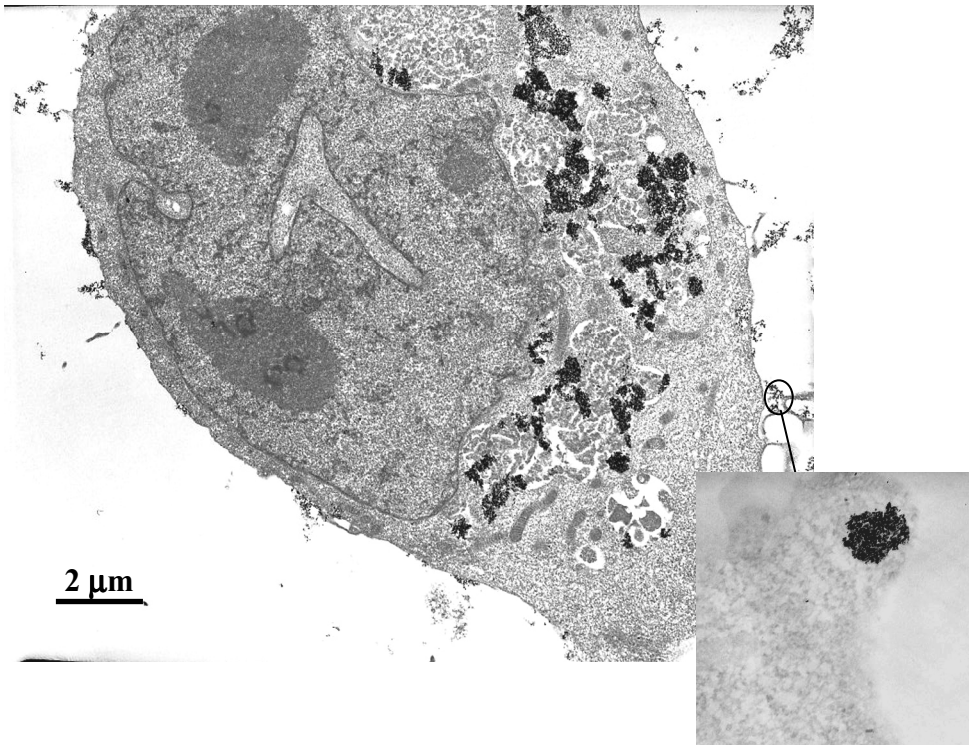

Figure S6. Transmission electron microscopy image of mammal cell (MDA-MB-231) incubated with HMBP coated nanocrystals for 48h showing nanoparticles incorporation into endocytotic vesicle.