Experimental Section

Materials. Perfluorooctyl iodide (C₈F₁₇I) was from Atochem, iron chloride hexahydrate (FeCl₃·6H₂O, 99% extra pure) from Acros Organics, undecenol, iron chloride tetrahydrate (FeCl₂·4H₂O, >99%), hydrochloric acid (HCl, min 37%), perfluorohexane (>99%) and Pluronic F68 (a polyoxyethylene-polyoxypropylene triblock copolymer, MW~8300, purity >99%) from Sigma Aldrich. Tetramethylammonium hydroxide (N(CH₃)₄OH aqua solution) was from Alfa Aesar. A HEPES buffer solution (N-2-hydroxyethylpiperazine-N’-2-ethanesulfonic acid, from Sigma) (20 mmol L⁻¹) in a 150 mmol L⁻¹ NaCl solution was prepared and its pH adjusted to 7.0 with 1N NaOH. Water was purified using a Millipore system (surface tension: 72.1 mN m⁻¹ at 20°C, resistivity: 18.2 MΩ cm).

Synthesis of (Perfluorooctyl)undecyl Phosphate C₈F₁₇(CH₂)₁₁OP(O)(OH)₂ (F8H11Phos). The non-commercially available precursor, C₈F₁₇(CH₂)₁₁OH, was prepared by radical addition of C₈F₁₇I on undecenol, followed by dehalogenation using gaseous HCl.¹ A solution of 130 mg (78 µL, 0.85 mmol, 2.5 eq) of POCl₃ in 1 mL of dry Et₂O was cooled to 0°C under argon and under stirring. 200 mg (0.34 mmol) of F8H11OH in 4 mL Et₂O were added dropwise over 60 min at 0°C and stirred at room temperature for 24 h, after which 0.5 mL of water was added and the mixture was stirred for 16 h. The two phases were separated and the aqueous phase was extracted with Et₂O (3 times). The combined ether phases were extracted with a 5% NaHCO₃ solution (3 times). The pH of the aqueous phase was adjusted to ~1 with diluted HCl and extracted with ethyl acetate (3 times). The organic phase was
washed with water, dried over MgSO₄ and the solvent evaporated to yield 190 mg (84%) of F₈H₁₁Phos (white solid). \(^1\)H NMR (CD₃OD, 400 MHz) \(\delta\) (ppm): 1.33-1.43 (m, 14H), 1.55-1.75 (m, 4H), 2.15 (m, 2H), 3.96 (dt, J = 6.8, 6.6 Hz). \(^{13}\)C NMR (CD₃OD, 75 MHz) \(\delta\) (ppm): 21.3, 26.7, 30.1, 30.3, 30.4, 30.5, 30.6, 30.7, 31.5, 31.7, 67.8. Anal. Calcd. for C₁₉H₂₄F₁₇O₄P: C, 34.04; H, 3.61. Found: C, 34.4; H, 3.82.

**Synthesis of Fe₃O₄ Nanoparticles.** Solutions of FeCl₃.6H₂O (1 mol L\(^{-1}\)) and FeCl₂.4H₂O (2 mol L\(^{-1}\)) were prepared by dissolving the iron salts in HCl (2 mol L\(^{-1}\)) and deoxygenated with argon for 30 min. 10 mL of FeCl₃.6H₂O solution and 2.5 mL of FeCl₂.4H₂O solution were mixed and heated to 70°C under argon under stirring. A 1 mol L\(^{-1}\) N(CH₃)₄OH solution was then injected at 0.7 mL min\(^{-1}\) and vigorously stirred for 20 min. The precipitate was washed with distilled water until pH 7 was reached.

The nanoparticles’ size was 10 ± 1 nm, as determined by TEM.\(^{[2, 3]}\)

**Preparation of Bare Microbubbles.** F₈H₁₁Phos (10 mmol L\(^{-1}\)) was dispersed in the HEPES/NaCl buffer (one night, room temperature, magnetic stirring). Pluronic F68 was added to facilitate dispersion (F₈H₁₁Phos:F68 molar ratio 10:1). A 1 mL aliquot of F₈H₁₁Phos/F68 dispersion was transferred to a glass tube (inner diameter 18 mm, length 63 mm) and pre-sonicated under air at low power (setting 2) for 30 s at room temperature. The sonicator (Vibracell, Bioblock-Scientific, Illkirch, France) was operated at 20 kHz (3 mm titanium probe, output ~600 W, duty cycle 40%). The F₈H₁₁Phos/F68 dispersion was then sonicated for 15 s (setting 2, ~200 W) at room temperature under N\(_2\) saturated with perfluorohexane by bubbling through three successive perfluorohexane-containing vials. The sonicator probe was consistently placed 5 mm below the dispersion’s surface. The aggregated microbubble suspension (foam) was diluted with buffer (8 mL). Size fractionation was achieved by flotation under gravity (30 min). Aliquots (1 or 2 mL, in order to keep the initial values of the ultrasound attenuation coefficient comparable) were consistently sampled at a depth of 55 mm from the edge of the glass vial and injected into the buffer-filled ultrasonic measuring cell (cell volume: 140 mL) thermoregulated at 25°C.

**Preparation of Fe₃O₄ Nanoparticle-Decorated Microbubbles.** A 1 mL aliquot of the above F₈H₁₁Phos/F68 dispersion was placed in a glass tube with 1 mL of Fe₃O₄NP dispersion (5 mg mL\(^{-1}\)) in HEPES/NaCl buffer and vortexed. This mixture was sonicated for 15 s, or 2 min, under a perfluorohexane atmosphere and diluted with 8 mL of HEPES buffer. Size fractionation was achieved by flotation under gravity (30 min) and aliquots were injected in the ultrasonic cell for AAM. 30 min were allowed before analysis by OM and SLS.
Acoustical Determination of Microbubble Size Distributions. The attenuation of the acoustical pulse that propagates through the aqueous bubble dispersion was measured and fitted to standard simple-harmonic resonator curves to determine bubble sizes.[4-7] The setup has been designed so that the ultrasound field does not alter the stability of the bubbles: First, the acoustical power that is used is low (<0.1 W cm\(^{-2}\); peak-to-peak acoustical pressure <3 \(10^4\) Pa). Second, the wave package only lasts 10 \(\mu\)s, and only 256 data points were recorded during the whole experimental span time along a pseudologarithmic scale, ensuring that the bubbles were only weakly exposed to ultrasound.[8] Each measurement was repeated on 3 to 5 different bubble preparations.

Optical Microscopy. Three to four droplets of bubble dispersion were placed into a concave glass slide, covered with a glass slide, and observed with an Olympus BH2 (Tokyo, Japan) microscope. Rapid image acquisition used a Lumenera Infinity 2 CCD camera (Lumenera, Ottawa, Canada). Bubble radii were measured using the ImageJ software on 5 to 10 slides.[9]

Dynamic Light Scattering (DLS). A Malvern Zetasizer Nano ZS was used (scattering angle 90\(^\circ\)). The \(z\)-averaged hydrodynamic mean diameters of the Fe\(_3\)O\(_4\)NPs dispersed in the HEPES/NaCl buffer were determined using the Malvern software. The measurements were achieved at 25\(^\circ\)C on 5 \(10^{-3}\) mg mL\(^{-1}\) concentrated dispersions at pH 7.

Static Light Scattering (SLS). To obtain acceptable obscuration values, 1 mL of bubble dispersion prepared from a 10 mM-concentrated \(F8H11\)Phos dispersion was injected in the 10 mL HEPES buffer-containing cell of a Coulter LS100 instrument (Coulter Electronics Inc., Hialeah, FL). The samples were continuously agitated at 25\(^\circ\)C. Fraunhofer's theory was used to analyze the diffraction pattern.

**Figure S1.** Transmission electron micrograph (TEM) of a dried sample of Fe\(_3\)O\(_4\) nanoparticles. The average diameter of the individual magnetite core is 10 ± 2 nm.
**Figure S2.** Dynamic light scattering of Fe$_3$O$_4$ nanoparticles dispersed in the HEPES/NaCl buffer at pH 7. The 10 nm NPs form narrowly dispersed clusters of ~170 nm in diameter.

**Figure S3.** a) Size distributions at various time points of bare microbubbles stabilized by F8H11Phos in the HEPES buffer (pH 7), as determined by the acoustical attenuation method. b) Time evolution of the volume fraction of F8H11Phos bubbles, indicating a half-life of ~18 min.
**Figure S4.** Optical micrographs of Fe$_3$O$_4$NP-decorated MBs. a) In the absence of magnetic field $B$, the decorated microbubbles are distributed randomly. When submitted to the magnetic field, they string up and align horizontally (b) or vertically (c), depending on the direction of $B$. Inset: magnification of a trio of bubbles. A video of the experiment is available from the author.