Supporting Information

Experimental Section

All chemicals were purchased from Sigma-Aldrich (St. Louis, MO) and used without further purification except when mentioned specifically. The gold nanostar is synthesized following a previous paper.[18] 2 μM para-mercaptobenzoic acid (pMBA) was added followed by 1 μM mPEG-SH (MW 5000) incubation for 1 hour. Then, a thin layer of silica was coated on the nanoparticle by tetraethyl orthosilicate (TEOS) with ammonia as a catalyst. After thiol-functionalization with 3-mercaptopropyltrimethoxysilane (MPTMS) on the surface, the maleimido-mono-amide-DOTA from Macrocylics (Dallas, TX) was added at a molar excess of $2x10^6$ per nanoparticle from a stock solution of 5 mg ml^{-1}, reacted for 8 hours at room temperature and centrifuge washed (5000 g for 10 min) three times. The gadolinium chloride (10 mM) stock solution was added to the DOTA-activated nanoparticles with a ratio of Gd^{3+} to nanoparticle of $2x10^6$:1 in 10 mM MES buffer at pH 6.25 with 0.1% sodium dodecyl sulfate (SDS). The solution was heated for 4 hours and then centrifuge washed three times to remove all non-chelated Gd^{3+}.

Nanoparticle concentration, size distribution and ζ-potentials were measured with a NanoSight NS 500 (Amesbury, U.K.). Raman measurements were performed on a HORIBA Jobin Yvon LabRAM ARAMIS system (Edison, NJ) with 785 nm excitation (40 mW) and 10 s integration. Transmission electron microscopy (TEM) images were acquired on an FEI Tecnai G2 Twin transmission electron microscope (Hillsboro, OR) with 160 kV accelerating voltage. Two photon photo-luminescence imaging and photothermal therapy assessments were performed on an Olympus FV 1000 multiphoton microscope (Olympus America, PA), following the same

procedure mentioned in a previous paper by our group.[19] Cell samples were kept on a 37 °C heating stage and exposed to pulsed laser irradiation at 850 nm. Samples were scanned on a 1.5 T clinical whole body MR scanner (GE Healthcare, WI) with a standard quadrature birdcage head coil. An inversion recovery sequence (*TE*/*TR* = 9/5000 ms) with inversion times of 500 and 2,100 ms was used to calculate the T_1 relaxation times. CT images were acquired on a clinical CT scanner (GE Healthcare, WI) with submilimeter resolution. The BT549 breast cancer cells were obtained from Dr. Victoria Seewaldt as a gift. Cells were cultured in RPMI-1640 medium (Invitrogen, CA) containing 10% fetal bovine serum (FBS), 25 mM HEPES and 0.023 U ml^{-1} of insulin in an incubator with a humidified atmosphere (5% CO_2) according to the American Type Culture Collection (Manassas, Virginia) protocol. Cells in exponential growth phases were used to test cytotoxicity and uptake nanoprobes for tumor phantom preparation. BT549 cells were cultured with 0.1 nM nanoprobes in RPMI-1640 growth media for one day and fixed with 4% paraformaldehyde before tumor phantom preparation. The cytotoxicity from QMT nanoprobe was tested by Resazurin-based toxicology assay (TOX8) following experimental procedure mentioned in a previous study.[19] Cells (3000 cells per well) were seeded on 96-well plates for two days and then incubated with growth media containing QMT nanoprobes. Following nanoprobe incubation, each well was washed twice with phosphate buffered saline (PBS) solution and then filled with fresh media. TOX8 (10% v/v) was added and the plate was kept in the incubation for an additional 1 hour. Live cells reduce Resazurin (nonfluorescent) to Resafurin (fluorescent). The fluorescence intensity was measured on a plate reader (Fluostar Omega, Germany). The cell viability was calculated by the intensity ratio from wells treated with (test group) and without (control group) nanoprobes.

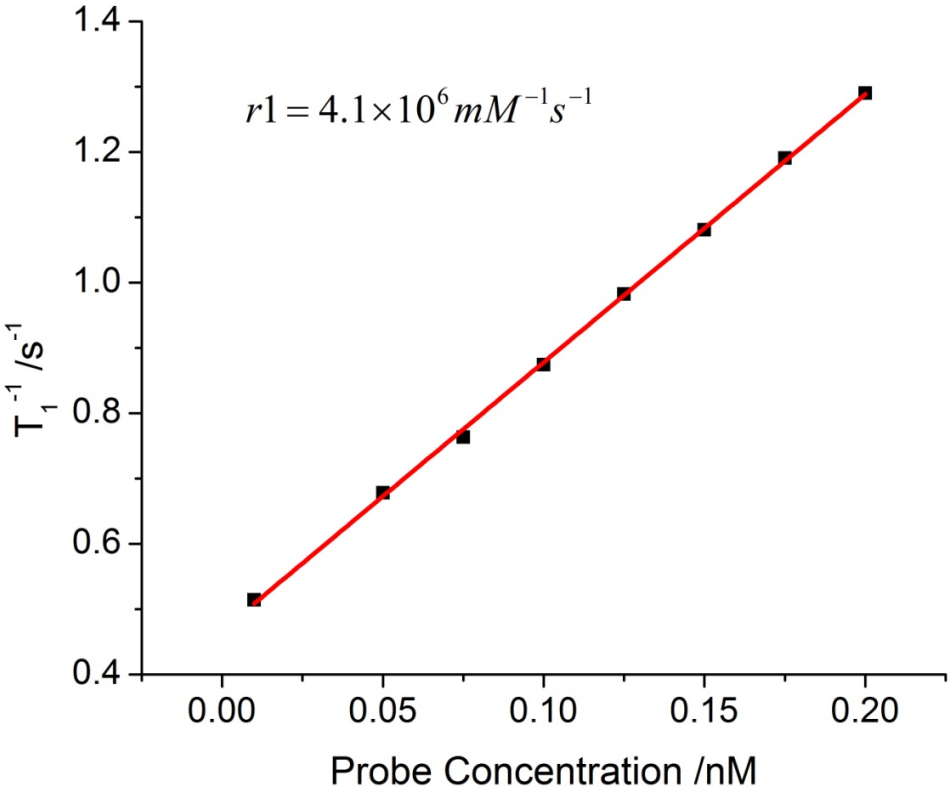

Figure S1. r1 relaxivity curve of developed QMT nanoprobes in water at 1.5 T magnetic field strength. The calculated r1 relaxivity is 4.1×10^6 mM^{-1} s^{-1}.

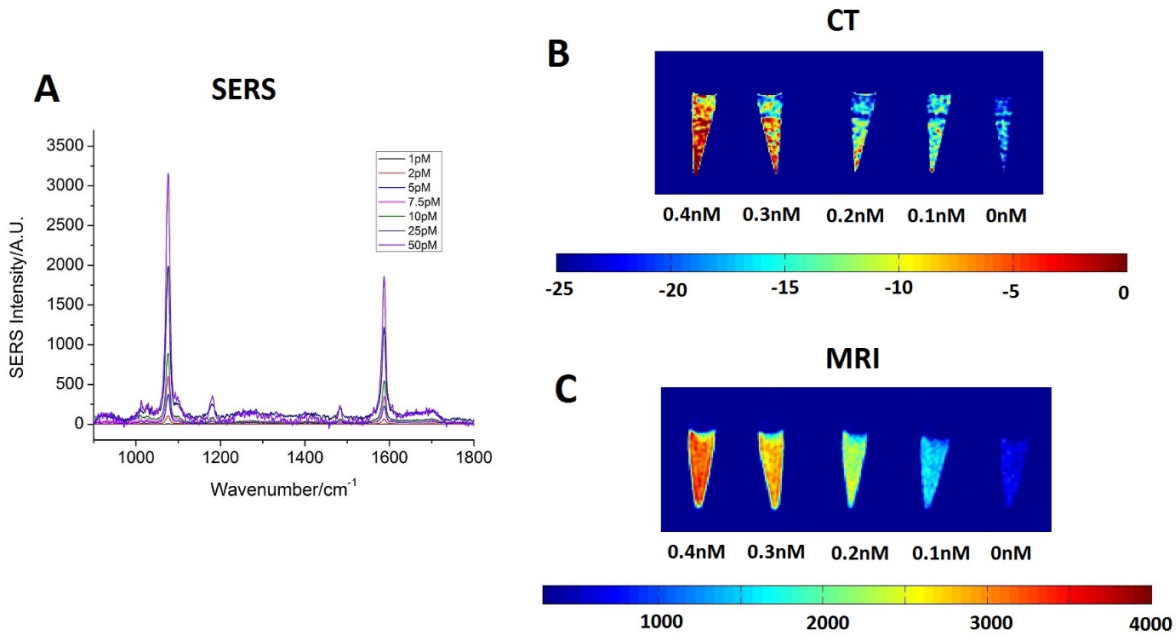

Figure S2. (A) SERS spectra of QMT nanoprobes with different concentration, (B) CT images of QMT nanoprobes with different concentration, (C) MRI images of QMT nanoprobes with different concentration.

Reference:

[18] A. M. Fales, H. Yuan, T. Vo-Dinh, *Langmuir* **2011**, *27*, 12186.
[19] H. Yuan, A. M. Fales, T. Vo-Dinh, *J. Am. Chem. Soc.* **2012**, *134*, 11358.