Supplementary Figure 1 | Results of magnetization measurements for the Fe(5)/TRGO, Fe(10)/TRGO, and Fe(20)/TRGO hybrids. (a) ZFC and FC magnetization curves of the Fe(5)/TRGO hybrid with two maxima identified in the ZFC magnetization curve. The maximum observed at a lower temperature (~26 K) corresponds to the magnetic blocking mechanism of the iron nanoparticle core with the most probable size in the system while the second maximum at
~55 K reflects the magnetic blocking of the nanoparticle surface shell of iron(III) oxide nature. (b) ZFC and FC magnetization curves of the Fe(10)/TRGO hybrid with one maximum at ~26 K related to the magnetic blocking of the iron nanoparticles. No other maximum is observed implying that Fe(III) shell is not completely developed and Fe(III) surface species undergo superparamagnetic/paramagnetic fluctuations even down to 5 K. (c) ZFC and FC magnetization curves of the Fe(20)/TRGO hybrid with profiles typical for bulk iron without any indication of transition to the superparamagnetic state.

Supplementary Figure 2 | HRTEM image of cavities (indicated by yellow arrows) in the TRGO matrix (imaged without the use of iron precursor) that serve as nucleation sites for the formation/growth of iron (metal) nanoparticles.
Supplementary Figure 3 | XPS spectra of the Fe(10)/TRGO hybrid. (a) C1s and (b) O1s XPS spectrum of the Fe(10)/TRGO hybrid.

Supplementary Figure 4 | Raman spectrum of the Fe(10)/TRGO nanocomposite.
Supplementary Figure 5 | Photographs of the Fe(10)/TRGO hybrid. Photographs of the Fe(10)/TRGO nanocomposite (a) before and (b) after placing a simple hand magnet (with an induction of ~0.3 T) against the wall of the box.
Supplementary Figure 6 | Chemical stability of the Fe(10)/TRGO hybrid under air conditions and in a physiological solution assessed by $^{57}$Fe Mössbauer spectroscopy. Room-temperature $^{57}$Fe Mössbauer spectra of (a) the freshly-prepared Fe(10)/TRGO hybrid after primary purification, (b) the hybrid after one month of storage under ambient laboratory conditions in the air, and (c) the hybrid after one month of storage in a physiological solution.
Supplementary Figure 7 | Colloidal stability of the Fe(10)/TRGO and Fe(10)/TRGO/PEG hybrid. Photographs comparing the colloidal stability of the freshly prepared Fe(10)/TRGO hybrid and a surface-modified Fe(10)/TRGO/PEG nanocomposite in physiological solution (left column) and a highly acidic (HCl) solution (right column). The concentration of the hybrid is 2 mg mL$^{-1}$ in each case, and the pictures show the suspensions as they appeared 1 hour after hybrid dispersion. The lower images demonstrate the drastically improved colloidal stability of the PEGylated system.
Supplementary Figure 8 | Optical microscopy image showing adherent non-degraded NIH/3T3 mouse fibroblast cells after 24 hours of incubation with 200 µg mL\(^{-1}\) of Fe(10)/TRGO/PEG hybrid.

Supplementary Figure 9 | In vitro MRI measurements. (a) The transverse relaxation rates (1/\(T_2\)) versus iron concentration for Fe(10)/TRGO (circles) and commercial FeraSpin XXL (squares) nanosystems with corresponding linear fittings (solid lines) measured in a field of 9.4 T. The values of \(r_2\) determined from these curves are 153 mM\(^{-1}\) s\(^{-1}\) for Fe(10wt.%)/TRGO and 64...
mM⁻¹ s⁻¹ for commercial FeraSpin XXL. (b) Plots of transverse relaxation rates (1/T₂* grey circles) and (1/T₂ black squares) for Fe(10)/TRGO nanocomposite with corresponding linear fittings (solid lines) measured under a field of 4.7 T. The values of r₂* and r₂ determined from these curves are 481.36 mM⁻¹ s⁻¹ and 163.25 mM⁻¹ s⁻¹, respectively. (c) Phantom experiments: comparison of negative contrast effect for FeraSpin XXL and Fe(10)/TRGO nanocomposite imaged by a 9.4 T MRI in T₂ weighted images and demonstrating remarkable differences in contrast properties.

Supplementary Figure 10 | Optical microscopy of Fe(10)/TRGO/PEG system before in vivo MRI application.
Supplementary Figure 11 | Results of magnetization measurements for the Co(10)/TRGO hybrid. (a) 5 K and 300 K hysteresis loops and (b) ZFC/FC magnetization curves recorded for the Co(10)/TRGO hybrid. The maximum in ZFC magnetization curve clearly demonstrating the passage of the Co nanoparticles from the high-temperature superparamagnetic state to the low-temperature magnetically blocked regime. The insets in panel a and b show behavior of the hysteresis loops of the Co(10)/TRGO hybrid around the origin and details of profile of the ZFC/FC magnetization curves of the Co(10)/TRGO hybrid at low temperatures, respectively.
Supplementary Figure 12 | Results of magnetization measurements for the Ni(5)/TRGO hybrid. (a) 5 K and 300 K hysteresis loops and (b) ZFC/FC magnetization curves recorded for the Ni(5)/TRGO hybrid. The maximum in ZFC magnetization curve clearly demonstrating the passage of the Ni nanoparticles from the high-temperature superparamagnetic state to the low-temperature magnetically blocked regime. The insets in panel a and b show behavior of the hysteresis loops of the Ni(5)/TRGO hybrid around the origin and details of profile of the ZFC/FC magnetization curves of the Ni(5)/TRGO hybrid at low temperatures, respectively.
Supplementary Table 1 | Mössbauer hyperfine parameter values. Mössbauer hyperfine parameter values determined by fitting the \(^{57}\)Fe Mössbauer spectra of the unfiltered and filtered Fe(10)/TRGO nanocomposite samples, where \(T\) is the temperature of the measurement, \(\delta\) is the isomer shift, \(\varepsilon_Q\) is the quadrupole shift (for doublet component), \(\Delta E_Q\) is the quadrupole splitting (for sextet component), \(B_{hf}\) is the hyperfine field, and RA is the relative spectral area of individual spectral components.

| Sample          | \(T\) (K) | Component | \(\delta\) ± 0.01 (mm s\(^{-1}\)) | \(\varepsilon_Q/\Delta E_Q\) ± 0.01 (mm s\(^{-1}\)) | \(B_{hf}\) ± 0.3 (T) | RA ± 1 (%) | Assignment         |
|-----------------|-----------|-----------|---------------------------------|--------------------------------|-----------------------|------------|---------------------|
| unfiltered      | 300       | Sextet    | 0.00                            | 0.00                           | 32.9                  | 31         | \(\alpha\)-Fe bulk |
| Fe(10)/TRGO     |           | Doublet   | 0.34                            | 0.70                           | -----                 | 27         | Iron oxide shell    |
|                 |           | Singlet   | 0.00                            | -----                          | -----                 | 42         | \(\alpha\)-Fe – SP |
| filtered        | 300       | Singlet   | 0.00                            | -----                          | -----                 | 79         | \(\alpha\)-Fe – SP |
| Fe(10)/TRGO     |           | Doublet   | 0.35                            | 0.71                           | -----                 | 21         | Fe\(^{3+}\) environment |
|                 | 50        | Sextet    | 0.08                            | 0.00                           | 33.1                  | 32         | \(\alpha\)-Fe – blocked |
|                 |           | Singlet   | 0.09                            | -----                          | -----                 | 46         | \(\alpha\)-Fe – SP |
|                 |           | Doublet   | 0.43                            | 0.75                           | -----                 | 22         | Fe\(^{3+}\) environment |
|                 | 5         | Sextet    | 0.11                            | 0.00                           | 33.2                  | 65         | \(\alpha\)-Fe – blocked |
|                 |           | Singlet   | 0.11                            | -----                          | -----                 | 13         | \(\alpha\)-Fe – SP |
|                 |           | Doublet   | 0.50                            | 0.72                           | -----                 | 22         | Fe\(^{3+}\) environment |

Supplementary Table 2 | Mössbauer hyperfine parameter values assessing the chemical stability of the Fe(10)/TRGO hybrid. Mössbauer hyperfine parameters derived from the room-temperature \(^{57}\)Fe Mössbauer spectra of the as-prepared Fe(10)/TRGO hybrid, the Fe(10)/TRGO hybrid after one month of storage under ambient laboratory conditions in the air, and the Fe(10)/TRGO hybrid after one month of storage in physiological solution. \(\delta\) is the isomer shift, \(\Delta E_Q\) is the quadrupole splitting, and RA is the relative spectral area of individual spectral components identified during fitting.

| Sample              | Component | \(\delta\) ± 0.01 (mm s\(^{-1}\)) | \(\Delta E_Q\) ± 0.01 (mm s\(^{-1}\)) | RA ± 1 (%) | Assignment      |
|---------------------|-----------|---------------------------------|---------------------------------|------------|----------------|
| as-prepared         | Singlet   | 0.00                            | 0.00                           | 79         | SP iron        |
|                     | Doublet   | 0.35                            | 0.71                           | 21         | Fe\(^{3+}\) species |
| one month in air    | Singlet   | 0.00                            | 0.00                           | 77         | SP iron        |
|                     | Doublet   | 0.34                            | 0.72                           | 23         | Fe\(^{3+}\) species |
| one month in        | Singlet   | 0.00                            | 0.00                           | 78         | SP iron        |
| physiological solution | Doublet   | 0.35                            | 0.69                           | 22         | Fe\(^{3+}\) species |
Supplementary Table 3 | Relaxivity indices ($r_1$ and $r_2$) of the most frequently used contrast agents under a field of 4.7 T. Data adopted from Rohrer, M., Bauer, H., Mintorovitch, J., Requardt, M. & Weinmann, H. J. Comparison of magnetic properties of MRI contrast media solutions under a field of 4.7 T. *Invest. Radiol.* **40**, 715-724 (2005).

| Trade name or international code | $r_1$ (mM$^{-1}$ s$^{-1}$) | $r_2$ (mM$^{-1}$ s$^{-1}$) | $r_2/r_1$ |
|----------------------------------|-----------------------------|-----------------------------|-----------|
| MAGNEVIST                        | 3.2 (3.0 – 3.4)             | 4.0 (3.8 – 4.2)             | 1.3       |
| GADOVIST                         | 3.2 (3.0 – 3.4)             | 3.9 (3.7 – 4.1)             | 1.2       |
| PROHANCE                         | 2.8 (2.7 – 2.9)             | 3.7 (3.5 – 3.9)             | 1.3       |
| MULTIHANCE                       | 4.0 (3.8 – 4.2)             | 5.0 (4.7 – 5.3)             | 1.3       |
| DOTAREM                          | 2.8 (2.7 – 2.9)             | 3.7 (3.5 – 3.9)             | 1.3       |
| OMNISCAN                         | 3.3 (3.1 – 3.5)             | 4.1 (3.9 – 4.3)             | 1.2       |
| TESLASCAN                        | 1.6 (1.5 – 1.7)             | 2.7 (2.6 – 2.8)             | 1.7       |
| OPTIMARK                         | 3.8 (3.5 – 4.1)             | 4.7 (4.5 – 4.9)             | 1.2       |
| RESOVIST                         | 2.8 (2.7 – 2.9)             | 176 (167 – 185)             | 62.9      |
| FERIDEX                          | 2.3 (2.2 – 2.4)             | 105 (100 – 110)             | 45.7      |
| GADOMER                          | 9.1 (8.6 – 9.6)             | 22 (21 – 23)                | 2.4       |
| MS-325                           | 5.5 (5.2 – 5.8)             | 6.9 (6.5 – 7.3)             | 1.3       |
| PRIMOVIST                        | 4.9 (4.7 – 5.1)             | 6.3 (6.0 – 6.6)             | 1.3       |
| SH U 555 C                       | 4.3 (4.1 – 4.5)             | 66 (63 – 69)                | 15.3      |