Electronic Supplementary Information

# Controlling Assembly and Spin Transport of Tetrathiafulvalene Carboxylate Coated Iron Oxide Nanoparticles

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## Supporting Figures and Tables

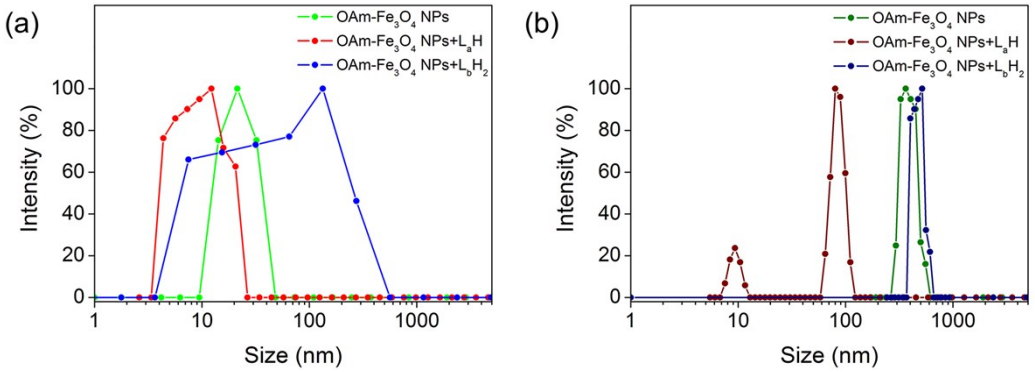


**Fig. S1.** DLS studies of OAm-$Fe_3O_4$ NPs, OAm-$Fe_3O_4$ NPs + $L_aH$, OAm-$Fe_3O_4$ NPs + $L_bH_2$ in (a) chloroform:ethanol (10:1/v:v) solvent, and (b) chloroform:ethanol (1:1/v:v) solvent. The concentration of these components (OAm-$Fe_3O_4$ NPs, $L_aH$, $L_bH_2$) are equal to what we used in the ligand exchange process.

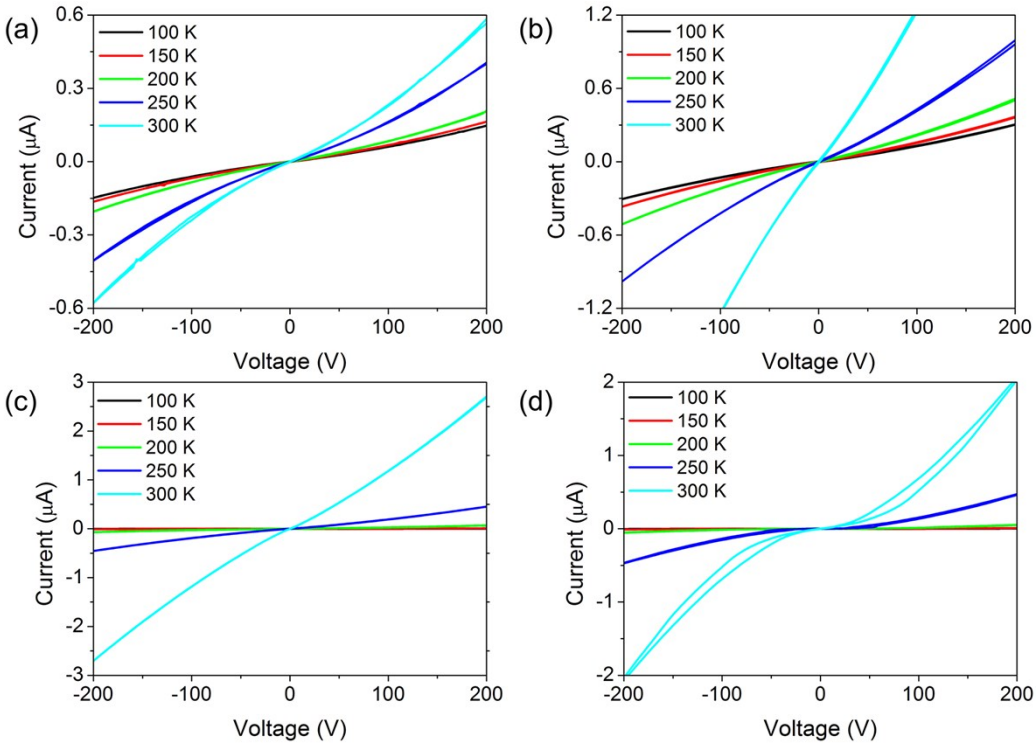


**Fig. S2.** *I*–*V* curves of (a) $L_a$-$Fe_3O_4$-10/1, (b) $L_a$-$Fe_3O_4$-1/1, (c) $L_b$-$Fe_3O_4$-10/1, and (d) $L_b$-$Fe_3O_4$ 1/1 NP assemblies at different temperatures.

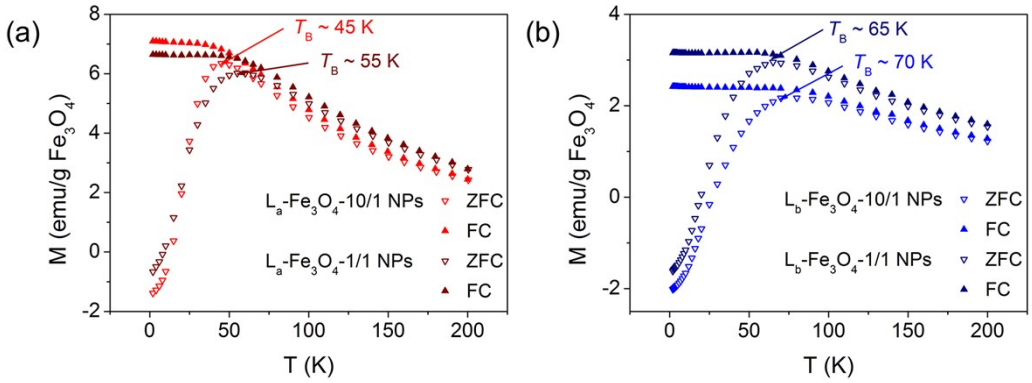


**Fig. S3.** ZFC-FC curves of (a) $L_a$-$Fe_3O_4$-10/1, (b) $L_a$-$Fe_3O_4$-1/1, (c) $L_b$-$Fe_3O_4$-10/1, and (d) $L_b$-$Fe_3O_4$-1/1 NP assemblies.

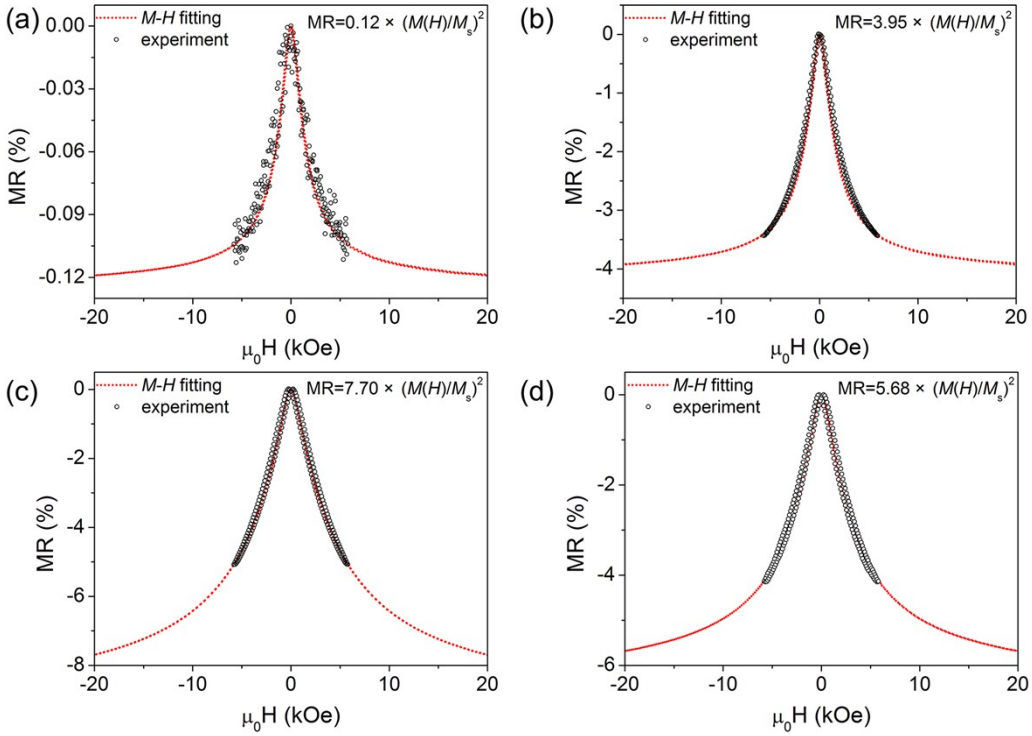


**Fig. S4.** Fitting MR-*H* curves and experimental MR-*H* scatterplots of (a) $L_a$-$Fe_3O_4$-10/1, (b) $L_a$-$Fe_3O_4$-1/1, (c) $L_b$-$Fe_3O_4$-10/1, and (d) $L_b$-$Fe_3O_4$-1/1 NP assemblies at 300 K. The $M_s$ in the inset equation equals to the *M* of the corresponding sample at 20 kOe, which can be obtained from Fig. 5.

**Table S1.** Mass percentage of Fe, $Fe_3O_4$ and organic layer[a] in the $L_a$- and $L_b$-coated $Fe_3O_4$ NPs and the grafting density[b] of the corresponding NPs.

| | *m*(Fe) (wt%) | *m*($Fe_3O_4$) (wt%) | *m*(L) (wt%) | *n* (1/nm$^2$)[b] |
|---|---|---|---|---|
| $L_a$-$Fe_3O_4$-10/1 | 56.6 | 78.1 | 21.9 | 1.30 |
| $L_a$-$Fe_3O_4$-1/1 | 59.2 | 81.8 | 18.2 | 1.03 |
| $L_b$-$Fe_3O_4$-10/1 | 57.0 | 78.7 | 21.3 | 1.27 |
| $L_b$-$Fe_3O_4$-1/1 | 57.9 | 80.0 | 20.0 | 1.18 |

a) The mass percentage of $Fe_3O_4$ is converted from the mass percentage of Fe determined by F-AAS. The mass of organic layer *m*(L) equales to $1 - m(Fe_3O_4)$.
b) The grafting density *n* is obtain by assuming a uniform 5.7 nm diameter and 5.2 g/cm$^3$ density for a sphere $Fe_3O_4$ core.

**Table S2.** Assignment of the main absorption bands[a] in the IR spectra of $L_aH$, $L_bH_2$ and the corresponding $Fe_3O_4$ NPs.

| mode assignment | $L_aH$ | $L_a$-$Fe_3O_4$ 10/1 NPs | $L_a$-$Fe_3O_4$ 1/1 NPs | $L_bH_2$ | $L_b$-$Fe_3O_4$ 10/1 NPs | $L_b$-$Fe_3O_4$ 1/1 NPs |
|---|---|---|---|---|---|---|
| *ν*C−H | 2955 m | 2957 m | 2955 m | 2956 m | 2957 m | 2955 m |
| | 2920 s | 2920 s | 2920 s | 2918 s | 2921 s | 2921 s |
| | 2851 s | 2850 s | 2851 m | 2850 s | 2851 s | 2851 s |
| *ν*C=O and | 1674 m | | | 1714 s | | |
| | | 1637 w | 1637 m | | 1618 m | 1619 m |
| *ν*C−O | 1294 m | | | 1304 s | | |
| | | 1391 s | 1379 m | | 1380 s | 1381 s |
| *ν*C=C | 1564 w | 1560 w | 1564 w | 1560 w | 1564 m | 1564 m |
| | 1531 w | 1530 w | 1533 w | 1550 w | 1536 m | 1536 m |
| *δ*$CH_2$ | 1466 w | 1459 w | 1460 w | 1470 s | 1462 m | 1462 m |
| *δ*O−H | 1418 s | | | 1421 w | | |
| *δ*$CH_3$ | 1381 w | ----[b] | ----[b] | 1361 w | ----[b] | ----[b] |
| | 1256 w | 1259 m | 1259 w | ----[b] | 1259 s | 1259 w |
| | | 1093 m | | | 1093 s | |
| | | 1020 m | | | 1020 s | |
| | | 802 m | | | 802 s | |
| *ν*S−C−S | 889 w | 886 w | 885 w | 890 w | 876 w | 887 w |
| Fe−O lattice | | 800−500 s,br | 800−500 s,br | | 800−500 s,br | 800−500 s,br |

a) Unit: cm$^{-1}$; s: strong; m: middle; w: weak; vw: very weak; br: broad.
b) Band that cannot be discerned due to the coverage of other bands.

**Table S3.** Atomic percentage[a)] of the main elements in $L_aH$ and $L_bH_2$ coated $Fe_3O_4$ NPs.

| | C | O | S | N | Fe |
|---|---|---|---|---|---|
| $L_a$-$Fe_3O_4$ 10/1 NPs | 51.99 | 28.18 | 5.47 | 0.00 | 14.36 |
| $L_a$-$Fe_3O_4$ 1/1 NPs | 43.42 | 32.63 | 5.51 | 0.00 | 18.45 |
| $L_b$-$Fe_3O_4$ 10/1 NPs | 48.70 | 26.40 | 5.44 | 0.00 | 19.46 |
| $L_b$-$Fe_3O_4$ 1/1 NPs | 47.41 | 26.57 | 6.38 | 0.00 | 19.64 |

[a)] Only C, O, S, N and Fe are included and the sum of their percentage is 100%.