Supporting Information for

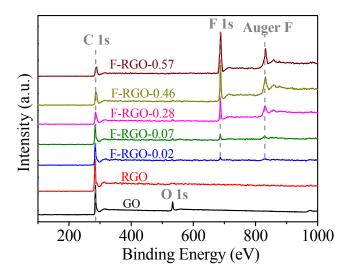
Obtaining High Localized Spin Magnetic Moments by Fluorination of Reduced Graphene Oxide

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XPS of purified GO, RGO, and the F-RGO samples.

Figure S1. XPS spectra of purified GO, RGO, and the F-RGO samples.

Figure S1 gives the XPS spectra of purified GO, RGO, and the F-RGO samples. One can find that the XPS spectrum of purified GO shows a dominant graphitic C 1s peak at 284.5 eV and O 1s

peak at ca. 532 eV. The C and O contents of purified GO are ca. 88.8 at. % and 11.2 at. %. After thermal reduction in Ar at 900 °C for 1 h, it is found that O 1s peak of RGO almost disappear, and the sample has a low content of oxygen (*ca.* 1.78 at. %). Note that the F 1s peak at ~688 eV and Auger F peak at ~833 eV of the F-RGO samples indicate the existence of C–F covalent bonding.

0.5 0.8 (b)(a) 0.4 0.6 W (emu/g) 0.4 J = 3/2J = 3/2(emu/g) 0.3 0.2 = 1 = 1 J = 1/2= 1/2F-RGO-0.28 F-GRO-0.46 RGO 0.2 0.1 0.0 0.0 15 20 H/T(kOe/K) 5 10 20 25 30 15 20 25 5 10 30 0 H/T(kOe/K)

Brillouin function fit for GO and the F-RGO samples.

Figure S2. Determination of the angular momentum quantum number *J* from M(H/T) curves. (a) The fit analysis of magnetization curves for RGO and F-RGO-0.28 using the Brillouin function for different *J* values. (b) Fits of the magnetization curve for F-RGO-0.46 using the Brillouin function for different *J* values. Symbols are experiment data and solid curves are fits to the Brillouin function for J=1/2, 1, 3/2.

To characterize the intrinsic magnetism of RGO and the F-RGO samples, the mass magnetization M of the samples (linear diamagnetic background subtracted) was plotted as a function of the reduced field H/T. The M(H/T) curves of RGO and the F-RGO samples are described by using the Brillouin function

$$M = M_{S} \left[\frac{2J+1}{2J} Coth \left(\frac{2J+1}{2J} x \right) - \frac{1}{2J} Coth \left(\frac{x}{2J} \right) \right]$$

where $M_s = NgJ\mu_B$, $x = gJ\mu_B H/(k_BT)$, g is the g-factor, J = L + S (*J* is total quantum number, *L* orbital angular quantum number, and *S* spin quantum number) and *N* is the number of magnetons. Assuming g = 2, one can find that the Brillouin function provides excellent fits for all the samples. For example, Figure S2a gives the fit analysis of magnetization curves for RGO and F-RGO-0.28, and it is observed that the Brillouin function provides excellent fits for J = S = 1/2. For comparison, we also give fitting curves provided by J = 1, and 3/2 Brillouin function, and all of which provide poor fits. Clearly, it indicates that the spin moment of single electron contributes to the magnetization of these samples. Actually, most of the FG samples expect the samples of F-RGO-0.42, F-RGO-0.46, and F-RGO-1 show such spin-half paramagnetism. However, for F-RGO-0.46 the Brillouin function function provides good fits only using J = 1, with other values of *J* giving poor fits [Figure S2b].

The contents of the metal impurity elements of RGO and the F-RGO samples.

Table S1. The contents of the metal impurity elements (such as Fe, Co, Ni, Mn or Al) of all the samples measured by the ICP spectrometry. The unit is ppm, and 'ND' denotes 'not found'.

samples	Fe	Co	Ni	Mn	Al
RGO	8.0	ND	ND	2.0	ND
F-RGO-0.02	8.6	ND	ND	0.0	ND
F-RGO-0.07	11.0	ND	ND	0.0	ND
F-RGO-0.28	8.5	ND	ND	2.2	ND
F-RGO-0.42	2.0	ND	ND	2.0	ND
F-RGO-0.46	9.0	ND	ND	0.0	ND
F-RGO-0.52	6.0	ND	ND	2.0	ND
F-RGO-0.57	15.0	ND	ND	3.0	ND

F-RGO-1.0	8.0	ND	ND	0.0	ND