Supporting Information

Edge-Induced Room-Temperature Ferromagnetism in Carbon Nanosheets

Caixing Liu,^{abc} Yang Yang,^a Zongwei Ma,^a Chun Zhou,^a Dayong Liu,^{*,d} Xuan Luo,^d Xuebin Zhu,^d Yuping Sun,^d Zhigao Sheng^{*,ac}

^a Anhui Province Key Laboratory of Condensed Matter Physics at Extreme Conditions, High Magnetic Field Laboratory, Chinese Academy of Science, Hefei 230031, China

^b Hefei National Laboratory for Physical Sciences at Microscale, University of Science and Technology of China, Hefei 230026, China

^c Key Laboratory of Photovoltaic and Energy Conservation Materials, Chinese Academy of Sciences, Hefei, 230031, China

^d Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China

Table of Contents

Calculations for crystallite size or interedge distance	S3
The fitting of PM component by Brillouin function	S4
The computation details	S4
Figure S1. TEM images of CNs	S5
Figure S2. XRD, EDS and XPS of the CNs sample	S6
Figure S3. <i>M-H</i> of different samples in repeated experiments	S7
Figure S4. Effect of water and air on ferromagnetism of CNs	S7
Figure S5. Raman spectra of CNs prepared with potassium in lower temperatur	eS8
Figure S6. <i>M-H</i> of CNs prepared with potassium in lower temperature	S8
Figure S7. TEM images of sample N1 to N4	S9
Figure S8. The diagrams of edges and magnetic configurations	S10
Table S1. ICP-MS of CNs	S10
Reference	S11

Calculations for crystallite size or interedge distance

Since 1970, Raman spectra were used to determine the crystallite size, corresponding to the average distance¹⁻² (L_D), after the work of Tuinstra and Koenig (TK) who have performed a systematic comparison between Raman spectra and X-ray data for a variety of sp² carbon materials.³ Whereas the latter has established for long the linear dependence of the A_D/A_G (the area ratio of D peak to G peak) on $1/L_D$ in the case of large L_D (whatever the excitation wavelength), it is clear that the power dependence of A_D/A_G is not constant for L_D smaller than 10 nm.^{1, 4-5} As a consequence, the TK law becomes approximate and cannot apply. For *La* below 10 nm, P. Mallet-Ladeira⁶ recommend using the half-width at half-maximum (HWHM, Γ) of the G band instead, which obeys the following correlation:

$$L_D$$
 (nm) = (68 - Γ_G) / 5.2

For samples N1-N4, the corresponding Γ is 30, 49, 60 and 64 cm⁻¹, respectively. So the corresponding L_D is 7.3, 3.6, 1.5 and 0.8 nm, respectively.

The fitting of PM component by Brillouin function

The Brillouin function is a special function that appears when studying the idealized paramagnetic (PM) signal of magnetic materials. The standard Brillouin function can be defined by the following equation:

$$M = Ng\mu_{\rm B}J\left[\frac{2J+1}{2J}\operatorname{coth}\left(\frac{2J+1}{2J}x\right) - \frac{1}{2J}\operatorname{coth}\left(\frac{1}{2J}x\right)\right]$$

where $x = gJ\mu_BH / k_BT$ and k_B is the Boltzmann constant. The g factor and the angular momentum number J define the initial slope of *M*-H whereas the saturation level depends on the number of present spins, *N*. Assuming g = 2 and J = 1/2, the Brillouin function provides excellent fits for the PM component of CNs (Figure 2d). And the N is about 1.03 × 10²⁰ g⁻¹.

Computation details

All calculations were performed by using the Vienna Ab-initio Simulation Package (VASP) based on the density functional theory (DFT). The generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) was used for the exchange-correlation potential. The projected augmented wave (PAW) methods were used, and the corresponding plane-wave energy cutoff was set to 400 eV. All the structural relaxations and self-consistent calculations including spin polarization were done until the forces and total energy converge to 20 meV/Å and 1.0×10^{-6} eV, respectively. The Brillouin zones were sampled with $2 \times 2 \times 1$ k-points for the small nanoflake and one Γ k-point for two large nanoflakes.



Figure S1. TEM images of CNs with two kinds of typical morphology.



Figure S2. (a) XRD pattern of CNs. (b) Fe 2p XPS spectrum of CNs. (c) EDS spectrum of CNs.



Figure S3. *M-H* loops of samples prepared in three repeated experiments.



Figure S4. (a) *M*-*H* loop of original CNs. (b) *M*-*H* loop of CNs immersed in water for one week.



Figure S5. Raman spectra of samples prepared with potassium metals in 210, 170 and 130 °C.



Figure S6. *M*-*H* loops of samples prepared with potassium metals in 210, 170 and 130 °C.



Figure S7. TEM images of sample N1 (a), N2 (b), N3 (c) and N4 (d). As illustrated in (a), the abundant long lattice fringes and obvious diffraction rings suggest that sample N1 has good crystallinity. As for (d), there is no detected fringe, which means that sample N4 is close to amorphous carbon film.



Figure S8. The diagrams of edges and magnetic configurations along the two edges. (a) zigzag edge. (b) armchair edge. (c) The magnetic configurations of edges. As shown in the above enlarged image, the magnetic moments along armchair edge are tiny. And the armchair edge displays non-magnetic state due to the antiferromagnetic correlation between the adjacent edge atoms, which is consistent with the previous studies.⁷ Area of every circle is proportional to the magnitude of the magnetic moment at every atom.

Elements	Mn (ppm)	Fe (ppm)	Co (ppm)	Ni (ppm)
CNs	7.96	46.34	< 0.01	< 0.01

Table S1. Concentrations of magnetic elements in CNs, as determined by ICP-MS.

Reference:

(1). Ferrari, A. C.; Basko, D. M., Raman Spectroscopy as a Versatile Tool for Studying the Properties of Graphene. *Nat. Nanotechnol.* **2013**, *8*, 235-246.

(2). Ferrari, A. C., Raman Spectroscopy of Graphene and Graphite: Disorder, Electron-Phonon Coupling, Doping and Nonadiabatic Effects. *Solid State Commun.* **2007**, *143*, 47-57.

(3). Tuinstra, F.; Koenig, J. L., Raman Spectrum of Graphite. J. Chem. Phys. 1970, 53, 1126-&.

(4). Cançado, L. G.; Takai, K.; Enoki, T.; Endo, M.; Kim, Y. A.; Mizusaki, H.; Jorio, A.; Coelho, L. N.; Magalhães-Paniago, R.; Pimenta, M. A., General Equation for the Determination of the Crystallite Size La of Nanographite by Raman Spectroscopy. *Appl. Phys. Lett.* **2006**, *88*, 163106.

(5). Cancado, L. G.; Jorio, A.; Ferreira, E. H. M.; Stavale, F.; Achete, C. A.; Capaz, R. B.; Moutinho, M.
V. O.; Lombardo, A.; Kulmala, T. S.; Ferrari, A. C., Quantifying Defects in Graphene Via Raman Spectroscopy at Different Excitation Energies. *Nano Lett.* **2011**, *11*, 3190-3196.

(6). Mallet-Ladeira, P.; Puech, P.; Toulouse, C.; Cazayous, M.; Ratel-Ramond, N.; Weisbecker, P.; Vignoles, G. L.; Monthioux, M., A Raman Study to Obtain Crystallite Size of Carbon Materials: A Better Alternative to the Tuinstra-Koenig Law. *Carbon* **2014**, *80*, 629-639.

(7). Yazyev, O. V., Emergence of Magnetism in Graphene Materials and Nanostructures. *Rep. Prog. Phys.* **2010**, *73*, 056501.