

Supporting Information

Samples Characterization

Fig.S1 (Supporting Information) shows the typical XRD patterns of the obtained products with different average particle size 200nm (SR), 140nm (SG), 125nm (SB), all of the diffraction peaks could be easily indexed to iron oxides (JCPDS file 19-0629, magnetite, or JCPDS file 39-1346, maghemite), which matched well with our early report.¹ In the XRD pattern, no diffraction peaks corresponding to the carbon layer were observed because the coated carbon layer is amorphous. Meanwhile, the two peaks around 1585 and 1378 cm⁻¹ in the Raman spectrum (Figure S2) are assigned to the G and D mode of carbon, which indicates the existence of carbon in the sample.^{2,3}

Fig.S3 show the IR spectrum of the as-prepared samples, in which the two peaks around 1677 cm⁻¹ and 3400cm⁻¹ indicate the existence of carboxyl on the surface of the samples, so the samples exhibit negative zeta potential, which come from the negatively charged groups (carboxyl) on the surface of samples. The peak around 1590 cm⁻¹ is ascribed to C =C vibration, which also shows the possibility that the as-prepared CNPs contain carbon and the peak at 583 cm⁻¹ is originated to Fe—O stretching vibration.⁴ The magnetic hysteresis loop (Fig. S4) for the as-obtained samples was measured at room temperature (300 K) in an applied magnetic field of up to 5000 Oe, which indicates that these nanoparticles are superparamagnetic. All of the above evidences fully indicate that the prepared carbon-capped superparamagnetic colloidal nanoparticles have magnetite/carbon core/shell structure with the negatively charged groups (carboxyl) on the surface. So, the superparamagnetic colloidal

nanoparticles can be well re-dispersed in glycol under the intense sonication and remain stable in solution for at least several months.

Figure S6 shows the TEM images of the samples obtained by mixing superparamagnetic colloidal nanoparticles with two different kinds of average particle size under the induction of the magnetic field in a mass ratio of 1:1. In order to further observe the exact morphology of self-assembly structure in mixed color samples, we use the instant radical polymerization to fix the fast magnetically induced self-assembly of carbon-encapsulated superparamagnetic colloids with two different kinds of average particle size inside a polyacrylamide glycol gel matrix. The gel matrix was further observed as ultrathin sections microtomed using an Ultratome (Model MT-6000, Du Pont Company, USA) in liquid nitrogen. The ultrathin sections were transferred from liquid nitrogen to copper grids before observation with TEM to obtain the micrographs of the self-assembly structure of the superparamagnetic colloidal nanoparticles with two different kinds of average particle size (Fig. S7). These TEM photos show the magnetic particles with two different kinds of average particle size (SR+SB) forming double photonic band-gap hetero-structures and fixing inside the polymer matrix under the induction of the external magnetic field. Because the polymerization of the acrylamide monomer requires a variety of substances that involved in the reaction, so the polymerization process will inevitably have some negative impact on the self-assembly of the magnetic particles with two different kinds of average particle size and induce the corrosion of carbon layer.

References

- 1 H. Wang, Y. B. Sun, Q. W. Chen, Y. F. Yu, K. Cheng, *Dalton Trans.*, 2010, **39**, 9565–9569.
- 2 A. C. Ferrari, J. Robertson, *Phys. Rev. B: Condens. Matter*, 2000, **61**, 14095.
- 3 J. Ristein, R. T. Stief, L. Ley, W. Beyer, *J.Appl.Phys.*, 1998, **84**, 3836.
- 4 Q. Gao, *et al.* *J. Magn. Magn. Mater.*, 2009, **321**, 1052.

Figure captions:

Fig.S1. X-ray diffraction pattern of as-prepared carbon-capped superparamagnetic colloidal nanoparticles with different average particle size (SR) 200nm, (SG) 140nm, (SB) 125nm.

Fig.S2. Raman spectrum of the as-prepared carbon-capped superparamagnetic colloidal nanoparticles with different average particle size, (SR) 200nm, (SG) 140nm, (SB) 125nm at the range from 200 cm^{-1} to 1900 cm^{-1} .

Fig.S3. FT-IR spectrum of the as-prepared carbon-capped superparamagnetic colloidal nanoparticles with different average particle size, (SR) 200nm, (SG) 140nm, (SB) 125nm.

Fig.S4. The hysteresis loop is measured at room temperature (300 K) for obtained carbon-capped superparamagnetic colloidal nanoparticles with different average particle size (SR) 200nm, (SG) 140nm, (SB) 125nm in an applied magnetic field of up to 5000 Oe.,

Fig.S5. The solid (cyan) line represents the experimental reflection spectra of the new obtained optical diffraction color-cyan, the solid dotted lines (blue and green) show

the best-fitting profiles using two Gaussian functions, and the solid line (red) are the sum of the simulated distributions.

Fig.S6 Obtained TEM images of the double photonic band-gap hetero-structures formed under the induction of external magnetic field (a) (b) SR+SB, (c) (d) SR+SG and (e) (f) SG+SB.

Fig.S7 Obtained TEM observations of the ultrathin sections obtained from a cross-section along the magnetic field of the polyacrylamide glycol matrix, which further show the magnetic particles with two different kinds of average particle size forming double photonic band-gap hetero-structures under the induction of the external magnetic field (a, b, c, d) SR+SB.

Fig.S8 Obtained an optical microscopy image of the EG droplets that comprise the carbon-capped superparamagnetic colloidal nanoparticles in the PDMS matrix under a vertically aligned external magnetic field.

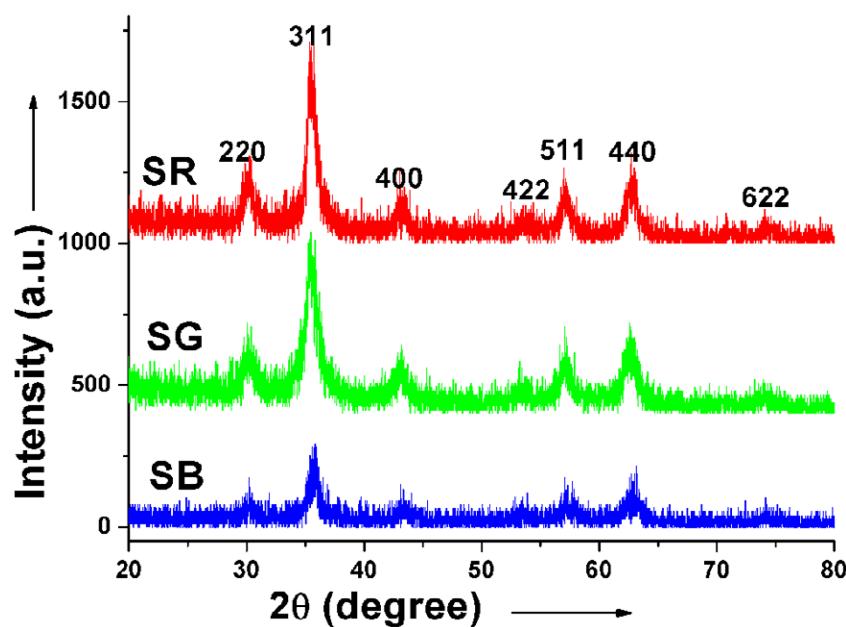


Fig.S1

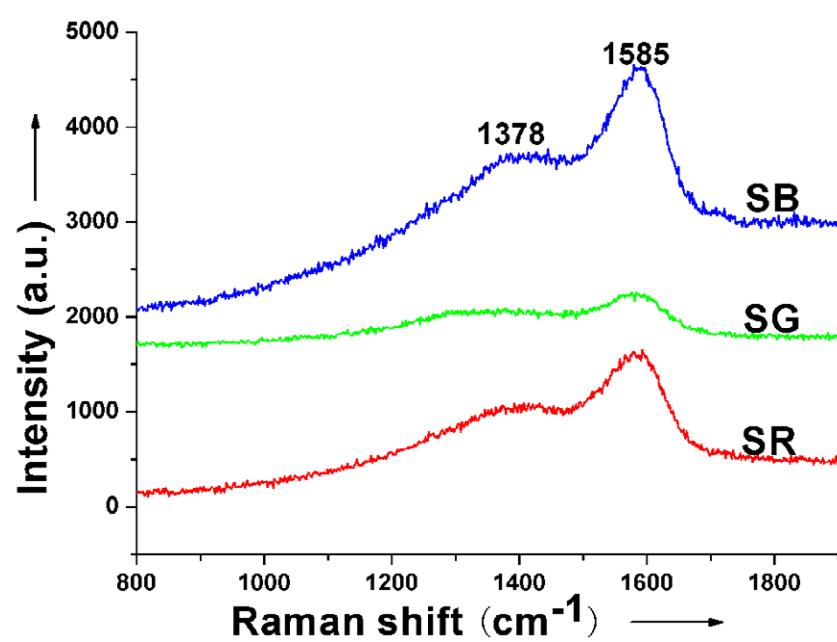


Fig.S2

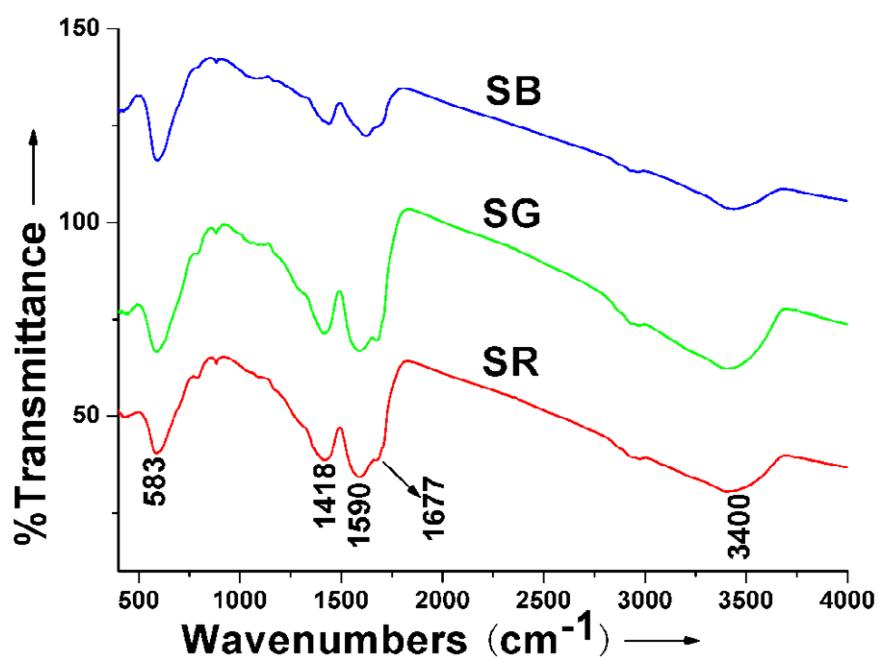


Fig.3

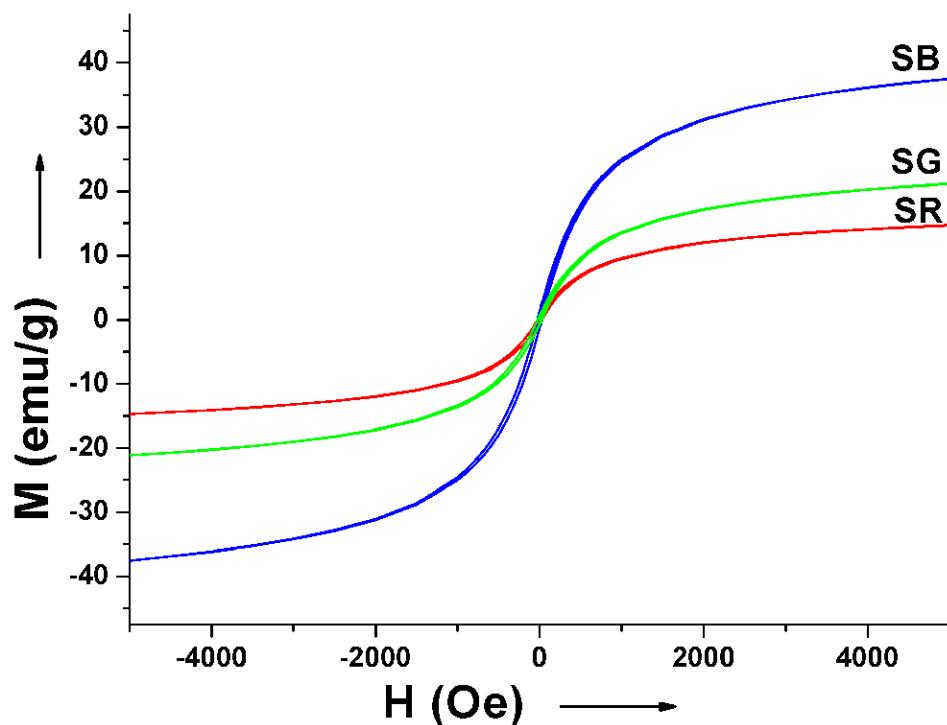


Fig.S4

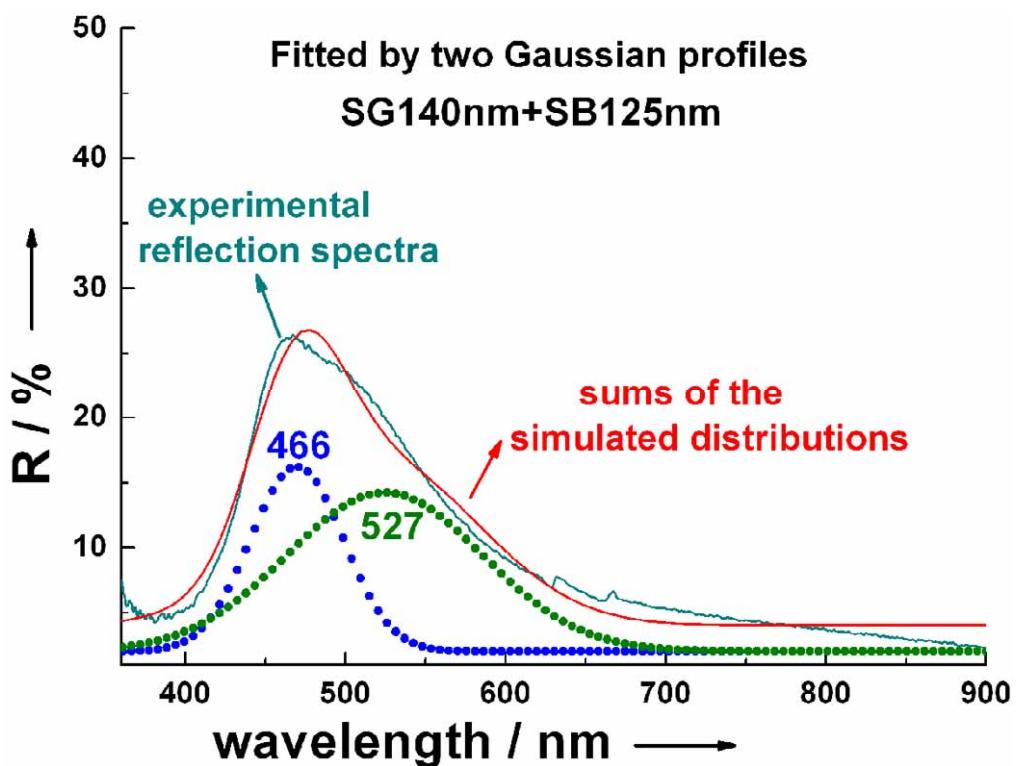


Fig.S5

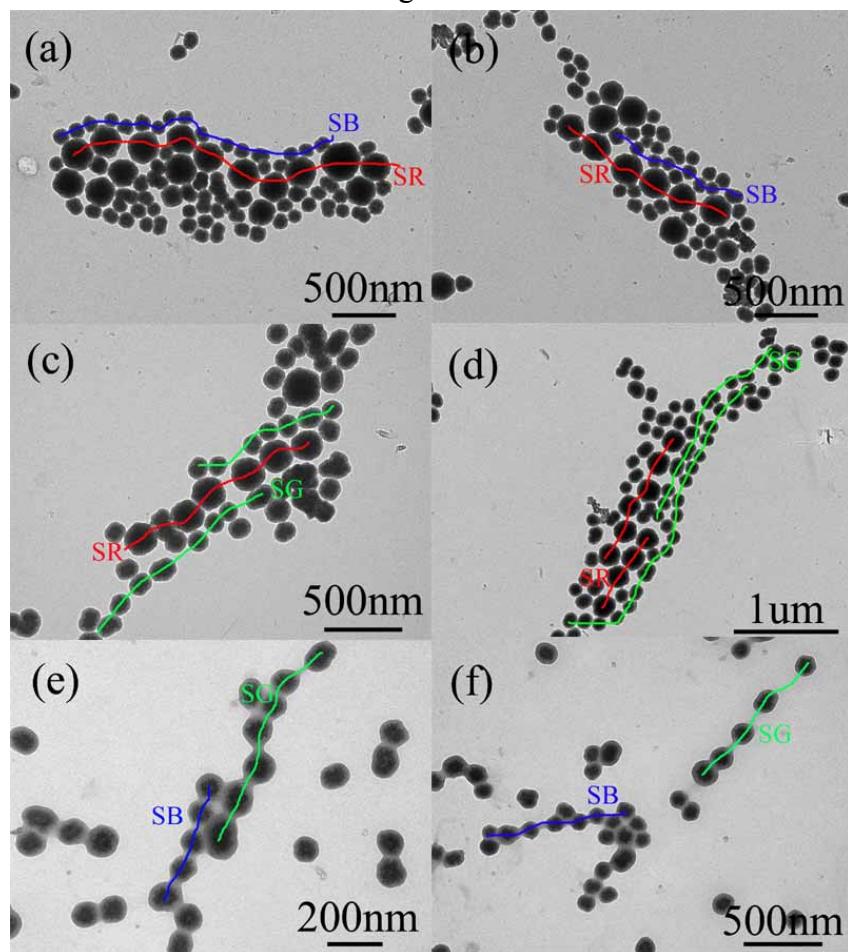


Fig.S6

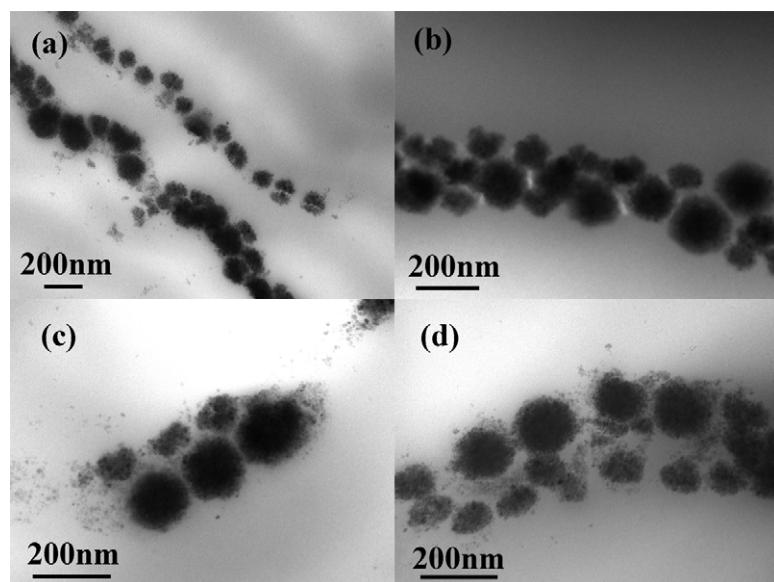


Fig. S7

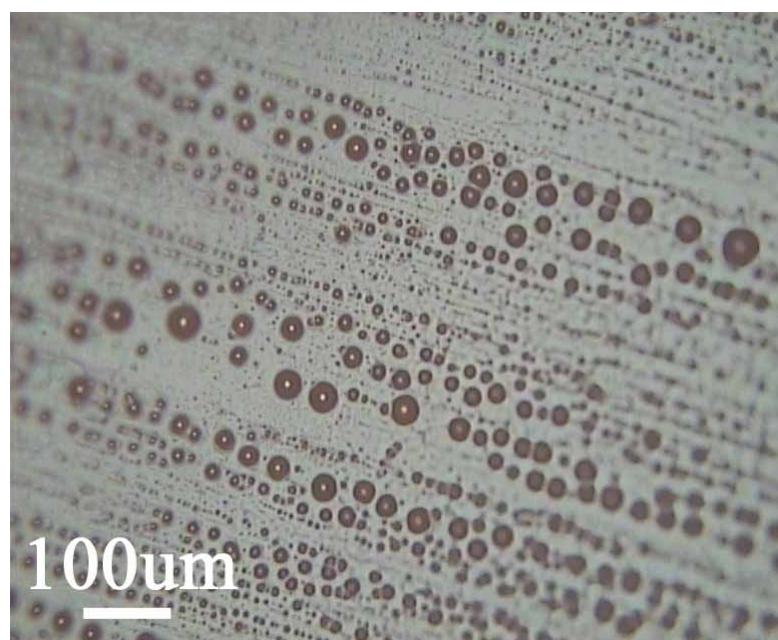


Fig.S8