Supplementary information

Dense Inter-particle Interaction Mediated Spontaneous Exchange Bias in NiO Nanoparticles

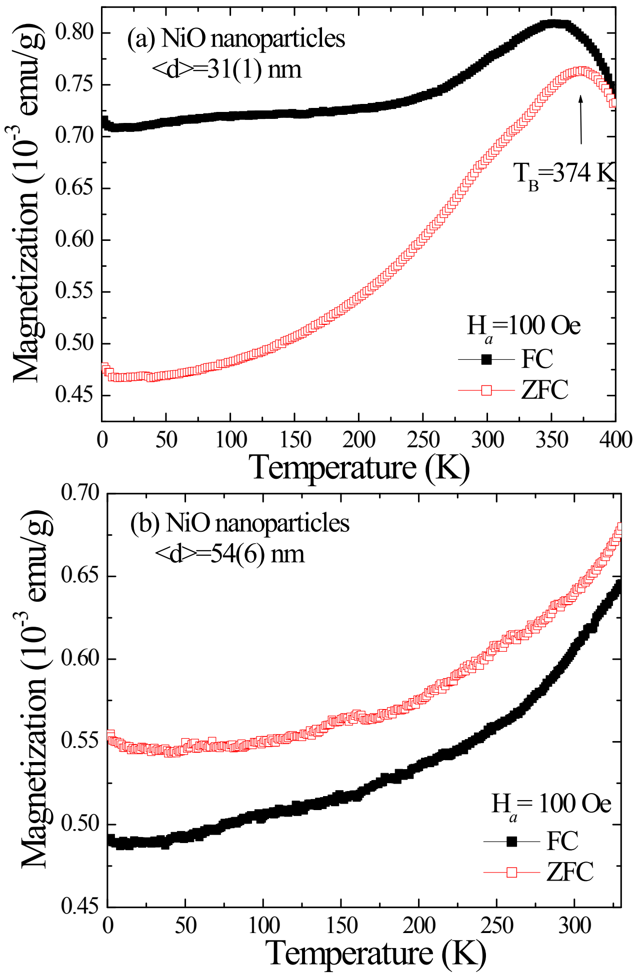
Ashish Chhaganal Gandhi[a,b], Jayashree Pant[c] , and Sheng Yun Wu[*,a]

[a]Department of Physics, National Dong Hwa University, Hualien 97401, Taiwan

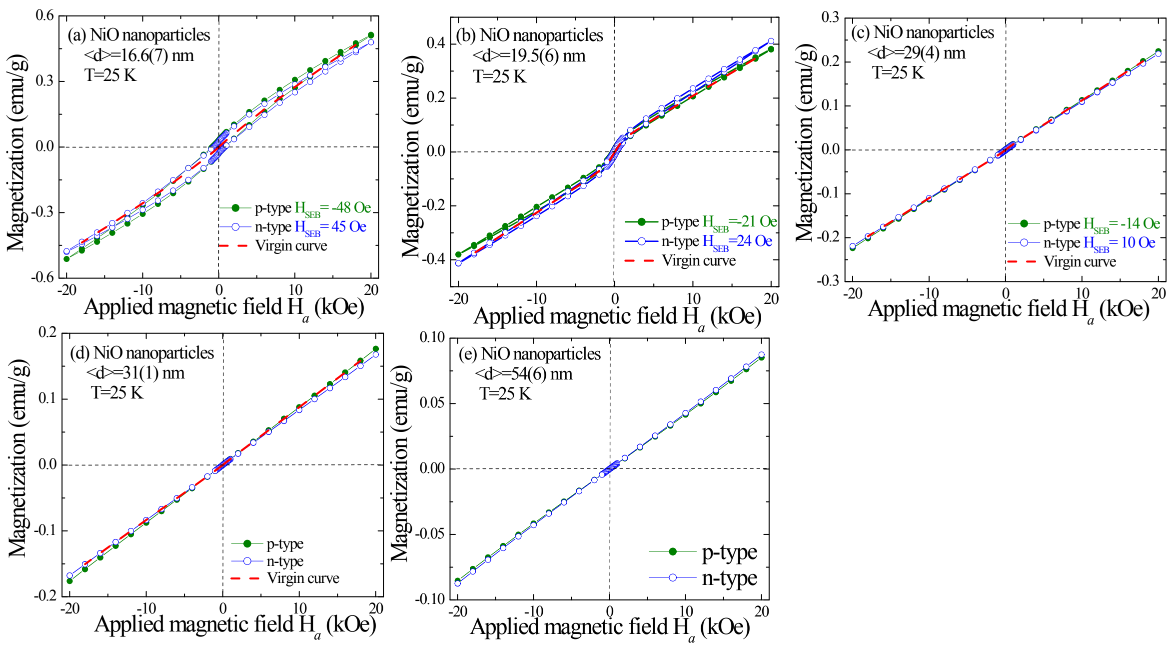
[b]Center for Condensed Matter Sciences, National Taiwan University, Taipei, Taiwan

[c]Department of Physics, Abasaheb Garware College, Savitribai Phule Pune University, India

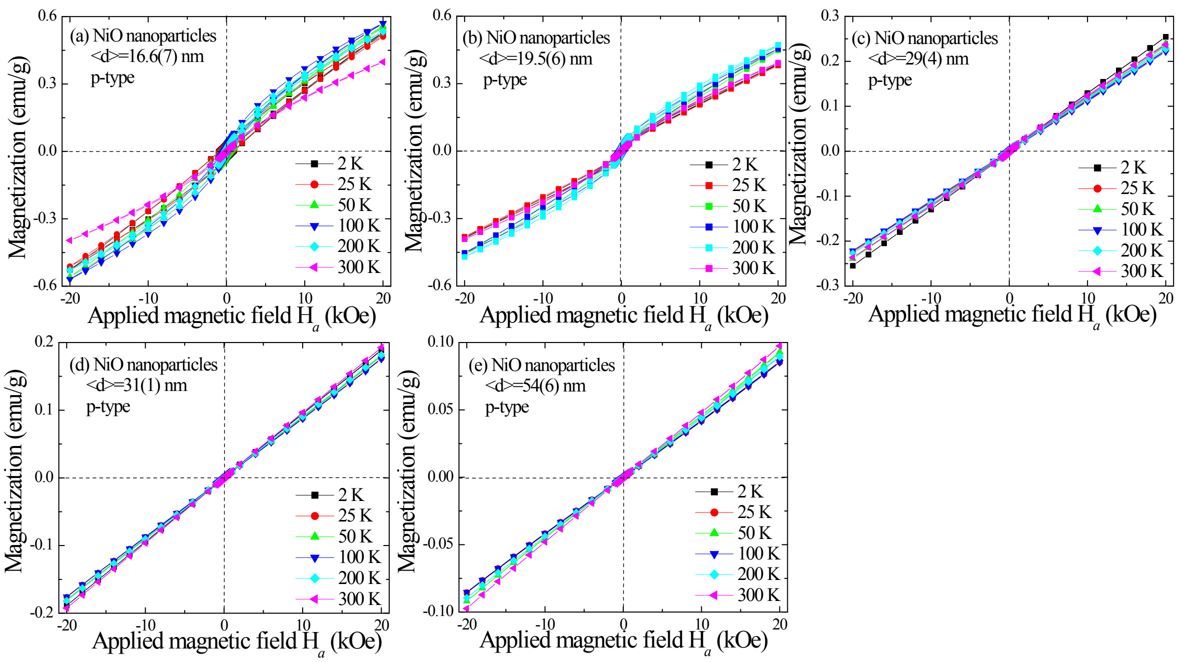
Figure S1 (a)-(b) M(T) curves measured with the ZFC-FC protocol in an external magnetic field of 100 Oe for 31(2) and 54(6) nm NiO nanoparticles, respectively



Figures S2 (a)-(e) show the full magnetization hysteresis $M(H_a)$ loops measured at 25 K with *p*- and *n*-type protocols after zero-field cooling for 16.6(7) to 54(6) nm NiO nanoparticles. The two-component magnetic behavior, ferromagnetic (FM) in the low field region and non-saturating antiferromagnetic (AFM) in the high field region is clearly visible in **Figures S2 (a)-(b)**[S2]. As the annealing temperature increases further the AFM component overcomes the FM component due to increased nanoparticle size and reduced nickel vacancies. Furthermore, the value of the magnetization observed at the highest applied field of 20 kOe decreases with the increase of particle size, which is in good agrement with previous reports[S3].



Figures S3 (a)-(e) Full magnetization hysteresis $M(H_a)$ loops measured at different temperatures of 2, 25, 50, 100, 200 and 300 K with the *p*-type protocol after zero-field cooling for 16.6(7) to 54(6) nm NiO nanoparticles.



Figures S4 (a)-(e) Full magnetization hysteresis *M*(*Ha*) loops measured at different temperatures of 2, 25, 50, 100, 200 and 300 K with the *n*-type protocol after zero-field cooling for 16.6(7) to 54(6) nm NiO nanoparticles.

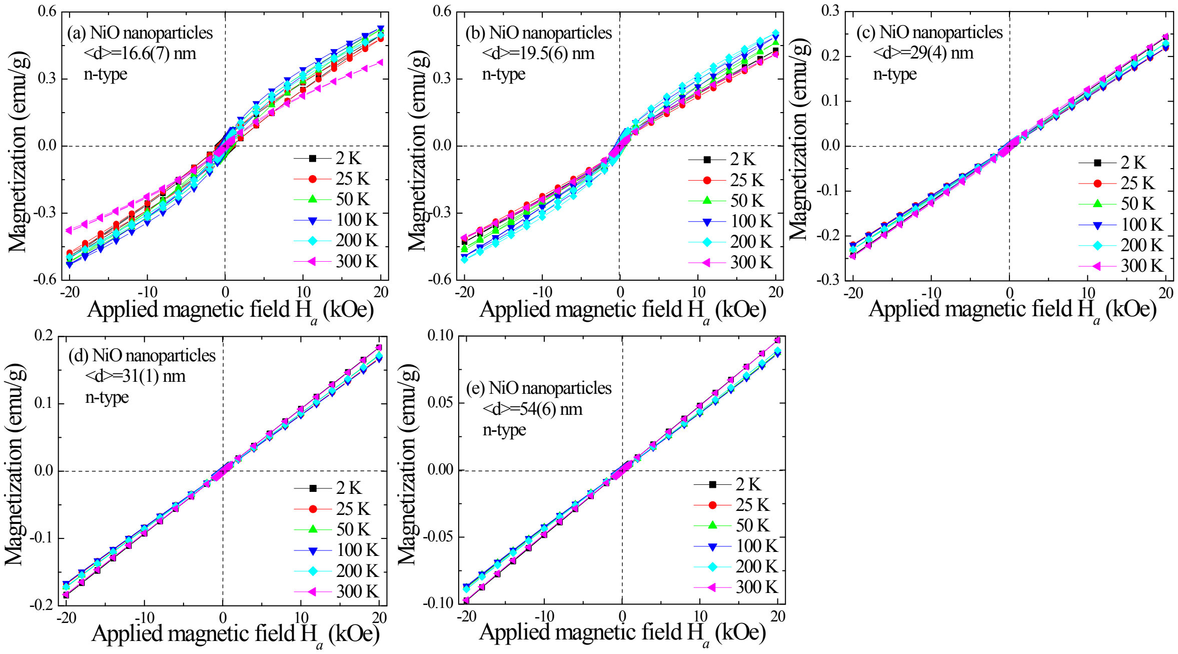
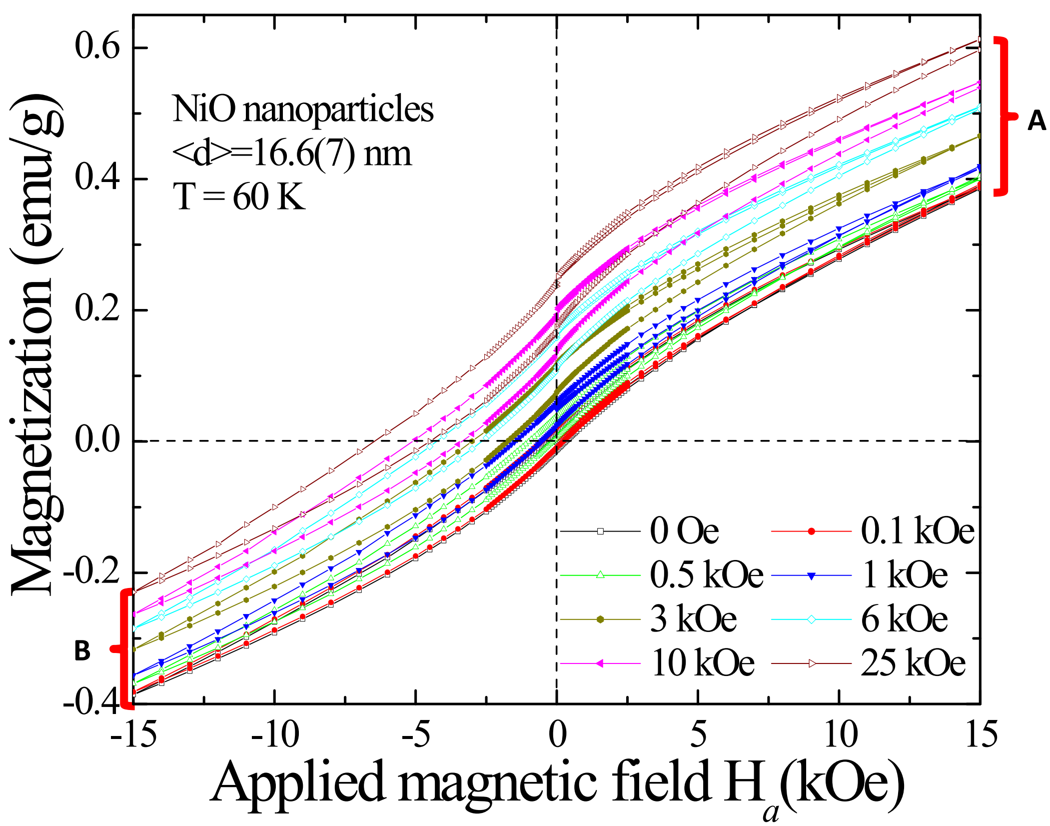


Figure S5 shows full magnetization hysteresis $M(H_a)$ loops measured at 60 K with the *p*-type protocol after field cooling (0 to 25 kOe) for 16.6(7) nm NiO nanoparticles. A and B indicate the observed asymeteric vertical loop shift in magnetization at higher magnetic fields.



References

S1. R. H. Kodama, A. E. Berkowitz, J. E. McNiff and S. Foner, *Phys. Rev. Lett.*1996, **77** 394-397

S2. S. Mandal, K. S. R. Menon, S. K. Mahatha, S. Banerjee, *Appl. Phys. Lett.* 2011, **99** 232507.

S3. S. D. Tiwari and K. P. Rajeev, *Phys. Rev. B* 2005,**72** 104433.

Table S1 Values of H_{SEB}, H_C, M_{VS}, and M_r obtained for *p*-type M(H_a).

Sample	Temperature (K)	H_{SEB} (Oe)	H_C (Oe)	M_{VS} (10^{-4}emu/g)	M_r (10^{-2}emu/g)
<d>=16.6(7) nm	2	-21	903	6.44	3.2511
	25	-47.5	827.5	13.94	3.0169
	50	-52.5	597.5	19.43	2.8228
	100	-37.5	376.5	18.97	2.2247
	200	-27	204	14.18	1.1407
	300	-24.5	79.5	8.25	0.2690
<d>=19.5(6) nm	2	-6.5	148.5	3.19	0.7839
	25	-20.5	197.5	9.97	1.0534
	50	-24.5	201.5	6.68	1.1696
	100	-16.5	259.5	9.22	1.5882
	200	-5.5	152.5	5.78	1.103
	300	-1.5	43.5	0.65	0.1709
<d>=29(4) nm	2	-3	50	-1.90	0.0831
	25	-14	62	3.00	0.1031
	50	-11.5	81.5	1.63	0.1297
	100	-24	143	3.55	0.2142
	200	-22	148	4.23	0.2414
	300	-32.5	88.5	5.25	0.151
<d>=31(1) nm	2	-15.5	6.5	1.19	0.0056
	25	-12	5	1.16	0.0048
	50	-11	14	1.09	0.0135
	100	-11	35	3.32	0.0104
	200	-7	36	3.64	0.0075
	300	2	12	-0.20	0.0089
<d>=54(6) nm	2	7.5	8.5	-0.31	0.0034

25	3	21	-0.13	0.0069
50	7	17	-0.35	0.0076
100	5	21	-0.14	0.0043
200	-5	18	0.25	0.0078
300	-4.5	22.5	0.097	0.0085

Table S2 Values of H_{SEB}, H_C, M_{VS}, and M_r obtained for *n*-type M(H_a).

Sample	Temperature (K)	H_{SEB} (Oe)	H_C (Oe)	M_{VS} (10^{-4}emu/g)	M_r (10^{-2}emu/g)
<d>=16.6(7) nm	2	39	813	-12.17	2.8913
	25	45	734	-12.47	2.6788
	50	36	511	-12.28	2.4467
	100	13	301	-5.92	1.8637
	200	-4	165	1.55	0.9465
	300	-11	65	3.71	0.2091
<d>=19.5(6) nm	2	12.5	149.5	-3.02	0.3991
	25	24	194	-7.84	0.6610
	50	29.5	213.5	-9.94	0.8306
	100	35	256	-11.33	1.1282
	200	24	182	-6.83	0.8314
	300	3.5	45.5	-0.74	0.1237
<d>=29(4) nm	2	1.5	54.5	-1.9	0.0882
	25	10	63	-2.3	0.1085
	50	31	84	-2.8	0.1319
	100	23.5	147.5	-4.4	0.2197
	200	3	153	-3.0	0.2522
	300	29.5	82.5	-3.2	0.1564
<d>=31(1) nm	2	0	9	0.0064	0.0072
	25	-1.5	4.5	0.0951	0.0029
	50	-0.5	12.5	0.0807	0.0114
	100	-3	25	0.3718	0.0232
	200	0.5	26.5	0.0112	0.0258
	300	-5.5	9.5	0.3249	0.0057
<d>=54(6) nm	2	1	20	-0.0461	0.0103

25	-3	24	0.1720	0.0102
50	-2.5	13.5	0.1121	0.0054
100	-9	29	0.3051	0.0076
200	-5	17	0.2286	0.0068
300	-2	12	0.1236	0.0060