Supplementary Information

Room Temperature Ferromagnetic Ordering from Bound Magnetic Polarons in Rare Earth Doped Ultrathin MoS_2 Nanosheets

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Table S1. Determination of dopant volume and concentration

Dopant Conc. (%)	Stock Solution Conc. (mM)	Vol. of $ErCl_3$ solution added (mL)	Vol. of $TbCl_3$ solution added (mL)	Vol. of $EuCl_3$ solution added (mL)
2	2.5		0.2	
3	2.5	0.3	0.3	0.3
5	2.5	0.5	0.5	0.5
7	2.5		0.7	
8	2.5	0.8	0.8	0.8

$$(NH_4)_2MoS_4 \xrightarrow{\Delta\ (155\ ^oC - 280\ ^oC)} MoS_3 + 2NH_3 + H_2S$$

Since $(NH_4)_2MoS_4$ degrades to MoS_3 in 1:1 ratio, and the lanthanide rare earth ions (Er^{3+}, Tb^{3+}, Eu^{3+}) are expected to be substitutional dopants in the 2D nanosheet host lattices, the quantity of dopant solution added can be determined based upon molar ratio against the atomic proportion of Mo^{4+} sites in the system.

As representative, the calculations for 5% doping of Er^{3+}, Tb^{3+} and Eu^{3+} are as follows:

Since 0.025 mmol (6.67 mg) of $(NH_4)_2MoS_4$ produced 0.025 mmol of MoS_3 with an equivalent amount of Mo^{4+} sites.

For 5% Tb^{3+} doping, quantity of Tb^{3+} ions required = 0.025/100×5 = **0.00125 mmol**

For 2.5 mM of $TbCl_3$ solution, volume used in the reaction = 0.00125/2.5 = 0.0005 L = **500 µL**

Molar mass of $TbCl_3$ = 265.28 g/mol

To prepare the solution, mass of $TbCl_3$ salt dissolved in 1 mL deionized H_2O =
2.5 × 1/1000 × 265.28 = **0.6632 mg**

For 5% Er^{3+} doping, quantity of Er^{3+} ions required = 0.025/100×5 = **0.00125 mmol**

For 2.5 mM of $ErCl_3$ solution, volume used in the reaction = 0.00125/2.5 = 0.0005 L = **500 µL**

Molar mass of $ErCl_3$ = 273.62 g/mol

To prepare the solution, mass of $TbCl_3$ salt dissolved in 1 mL deionized H_2O =
2.5 × 1/1000 × 273.62 = **0.6841 mg**

For 5% Tb^{3+} doping, quantity of Tb^{3+} ions required = 0.025/100×5 = **0.00125 mmol**

For 2.5 mM of $TbCl_3$ solution, volume used in the reaction = 0.00125/2.5 = 0.0005 L = **500 µL**

Molar mass of $TbCl_3$ = 258.32 g/mol

To prepare the solution, mass of $TbCl_3$ salt dissolved in 1 mL deionized H_2O =
2.5 × 1/1000 × 258.32 = **0.6458 mg**

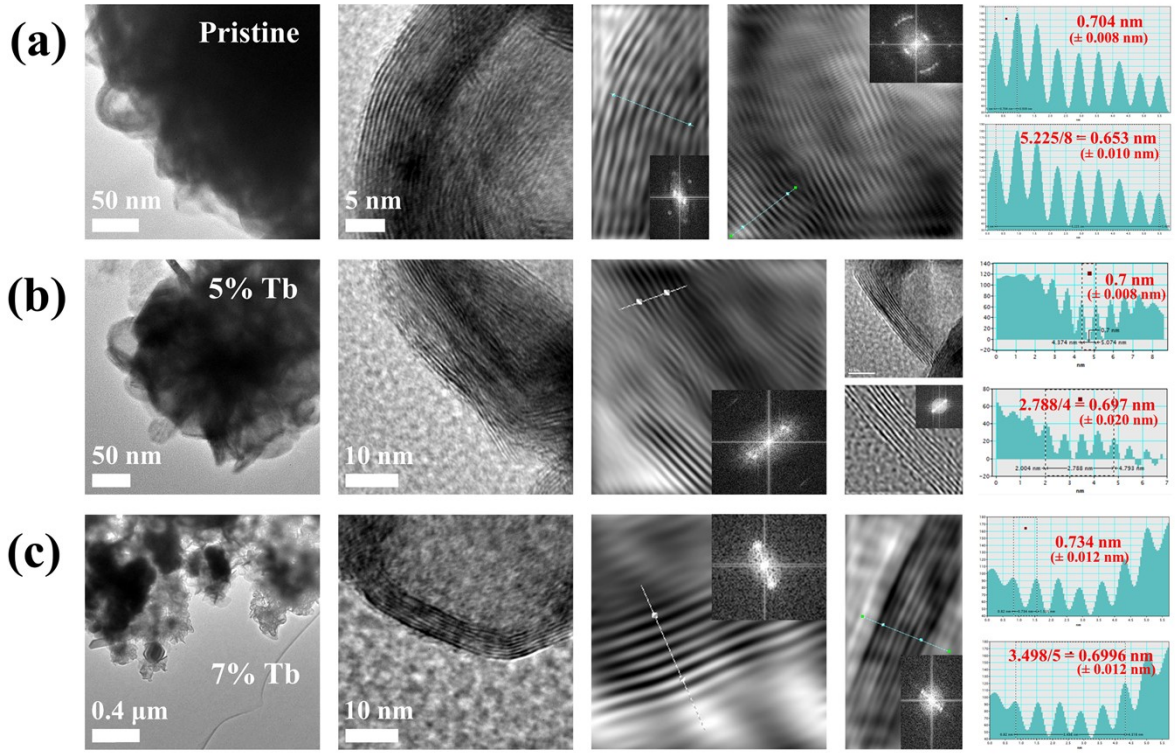


Figure S1. TEM and HRTEM characterizations for the MoS_2 nanosheets doped with varying Tb concentrations. (a) Pristine MoS_2 nanosheets (Left to Right): TEM, HRTEM, with corresponding SAED mapping and cross-sectional profile for the marked linear region. **(b)** 5% Tb-doped MoS_2 nanosheets (Left to Right): TEM, HRTEM, with corresponding SAED mapping and cross-sectional profile for the marked linear region. **(c)** 7% Tb-doped MoS_2 nanosheets (Left to Right): TEM, HRTEM, with corresponding SAED mapping and cross-sectional profile for the marked linear region.

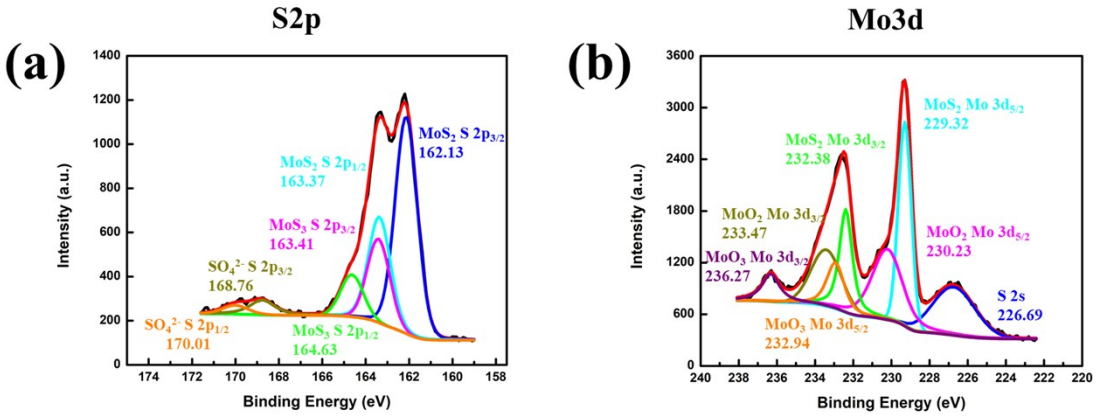
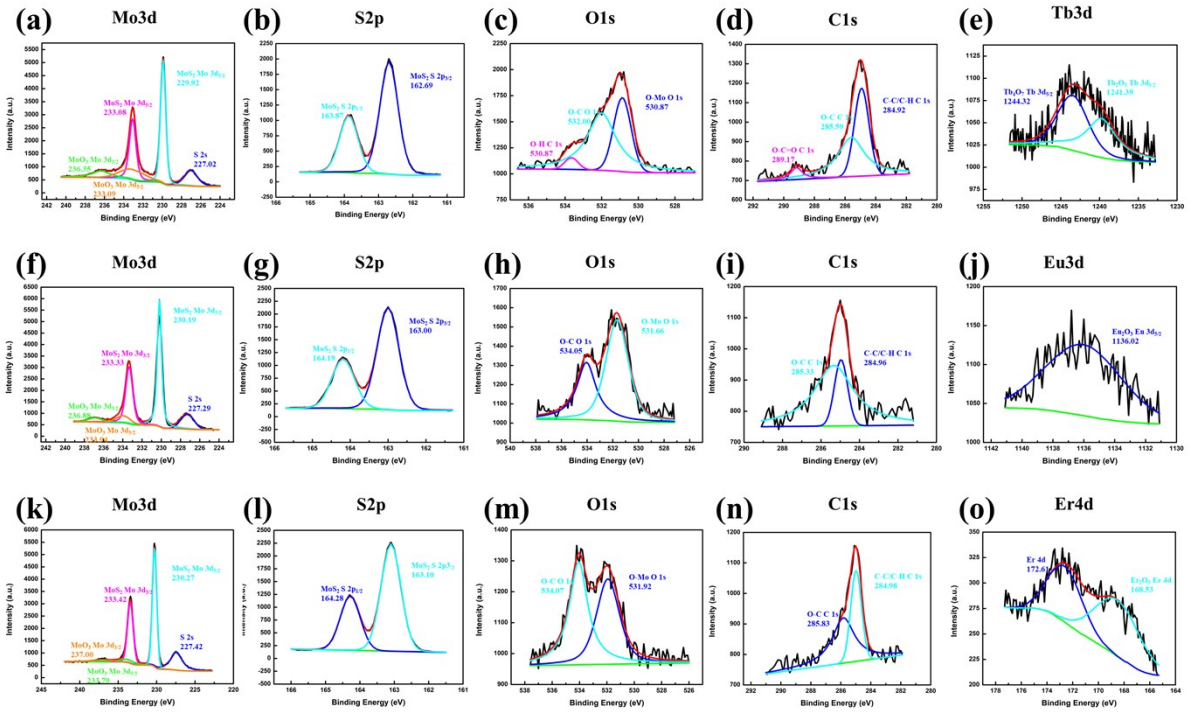


Figure S2. XPS characterization for pre-annealed MoS_3 nanosheets. (a) Signals pertaining to S2p elemental components, with distinct presence of MoS_3 labelled peaks (due to S_2^{2-} and bridging S contributions). **(b)** Signal pertaining to Mo3d elemental components, characterized by peak contributions from MoS_3 and MoO_x side product impurities.



(p) Tb^{3+}: MoS_2

Elements	Composition (At%)	
S $2p_{3/2}$ MoS_X	29.2	65.62
Mo $3d_{5/2}$ MoS_X	15.3	34.38
S/Mo	1.91	

(q) Eu^{3+}: MoS_2

Elements	Composition (At%)	
S $2p_{3/2}$ MoS_X	40.3	66.51
Mo $3d_{5/2}$ MoS_X	20.3	33.49
S/Mo	1.99	

(r) Er^{3+}: MoS_2

Elements	Composition (At%)	
S $2p_{3/2}$ MoS_X	42.7	68.32
Mo $3d_{5/2}$ MoS_X	19.8	31.68
S/Mo	2.16	

Figure S3. Full dataset of XPS characterization for MoS_2 nanosheets doped with different lanthanide rare-earth elements (Tb, Eu, Er). (a – e) MoS_2 nanosheets doped with 5% Tb, characterized of **(a)** Mo3d **(b)** S2p **(c)** O1s **(d)** C1s **(e)** Tb3d elemental signals. **(f – j)** MoS_2 nanosheets doped with 5% Eu, characterized of **(f)** Mo3d **(g)** S2p **(h)** O1s **(i)** C1s **(j)** Eu3d elemental signals. **(k – o)** MoS_2 nanosheets doped with 5% Er, characterized of **(f)** Mo3d **(g)** S2p **(h)** O1s **(i)** C1s **(j)** Er4d elemental signals. **(p – r)** Quantitative Mo:S atomic ratios for **(p)** Tb-doped MoS_2 **(q)** Eu-doped MoS_2 **(r)** Er-doped MoS_2 nanosheets.

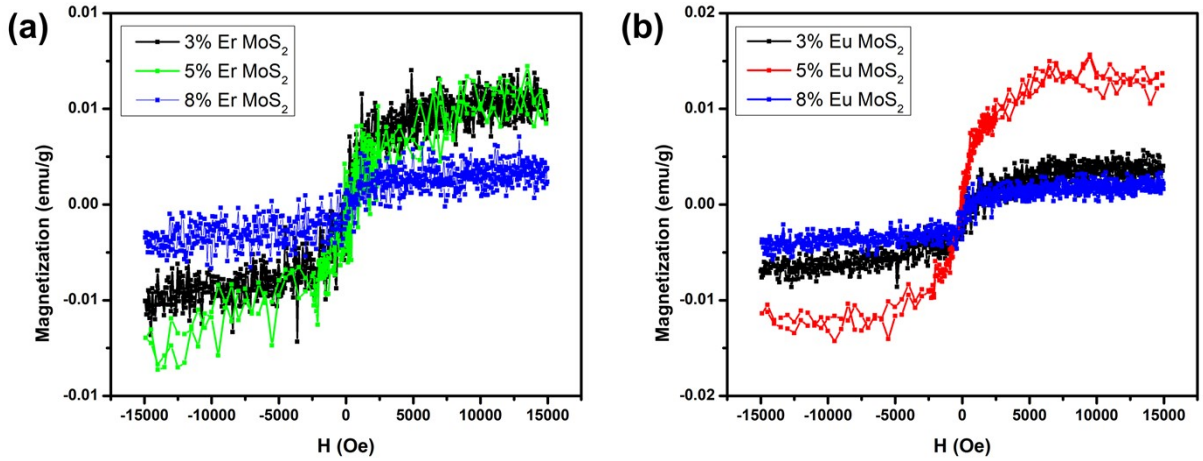


Figure S4. Magnetic characterization of Ln-doped MoS_2 (Ln = Er, Eu) nanosheets with variable atomic doping concentrations. (a) M-H plot for Er-doped MoS_2 nanosheets with 3%, 5% and 8% atomic Er^{3+} doping, after correction of background paramagnetic contributions. Trend comparison shows an optimal magnetization at 5% Er doping. **(b)** M-H plot for Eu-doped MoS_2 nanosheets with 3%, 5% and 8% atomic Eu^{3+} doping, after correction of background paramagnetic contributions. Trend comparison shows an optimal magnetization at 5% Eu doping.

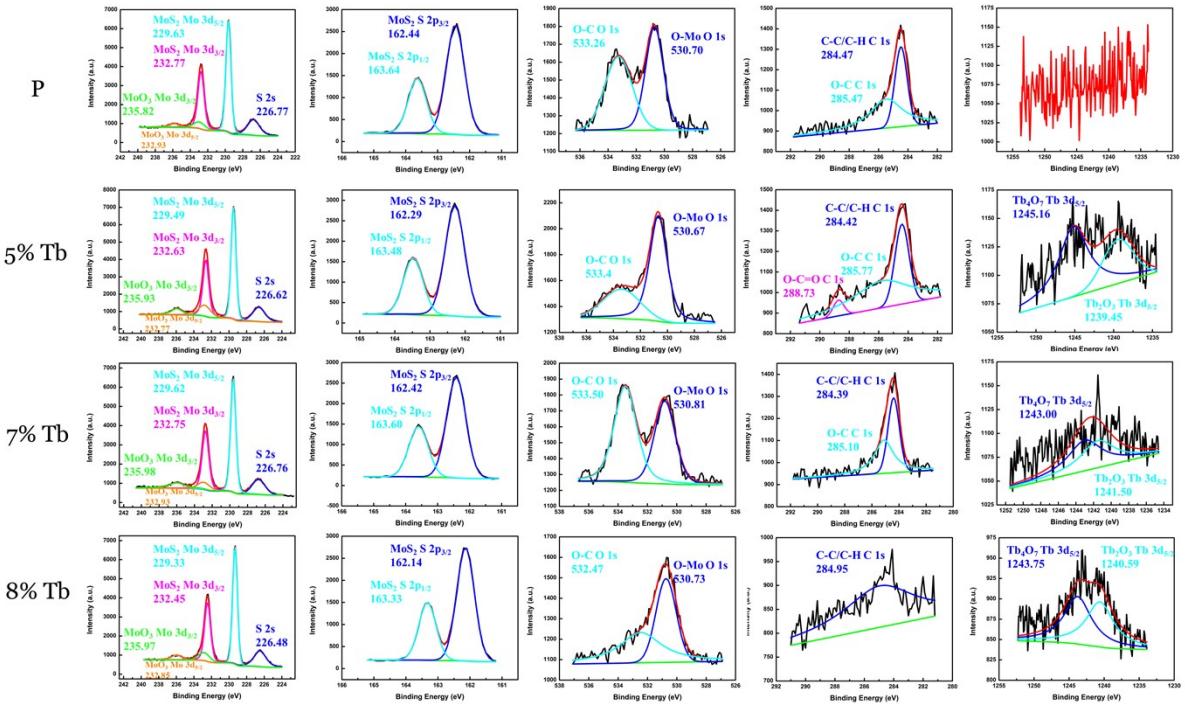
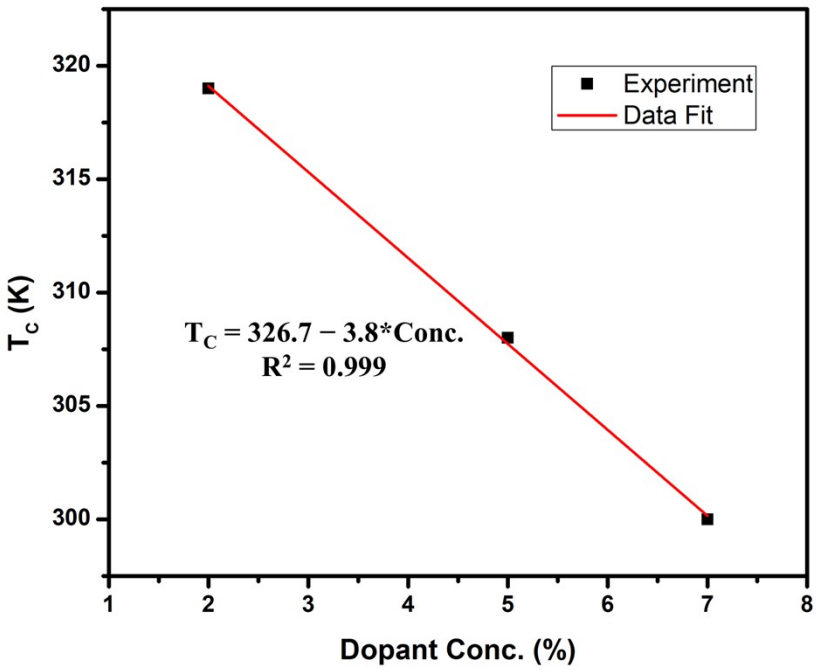


Figure S5. Full dataset of XPS characterization for Tb-doped MoS_2 nanosheets of various doping concentrations. (a) Pristine sample analyzed in terms of Mo3d, S2p, O1s, C1s and Tb3d elemental signals. **(b)** 5% Tb-doped MoS_2 sample analyzed in terms of Mo3d, S2p, O1s, C1s and Tb3d elemental signals. **(c)** 7% Tb-doped MoS_2 sample analyzed in terms of Mo3d, S2p, O1s, C1s and Tb3d elemental signals. **(d)** 8% Tb-doped MoS_2 sample analyzed in terms of Mo3d, S2p, O1s, C1s and Tb3d elemental signals.



	A	B	C	D
1	Model	Line		
2	Equation	y = A + B*x		
3	Reduced Chi-Sqr	0.10526		
4	Adj. R-Square	0.99884		
5			Value	Standard Error
6	Tc	A	326.68421	0.46483
7		B	-3.78947	0.09116

Figure S6. Linear correlation of T_C to dopant concentrations across the three sample measurements, with the associated fitting statistics, derived from temperature dependent FC-ZFC magnetic studies of 2% and 7% Tb-doped MoS_2 nanosheets.

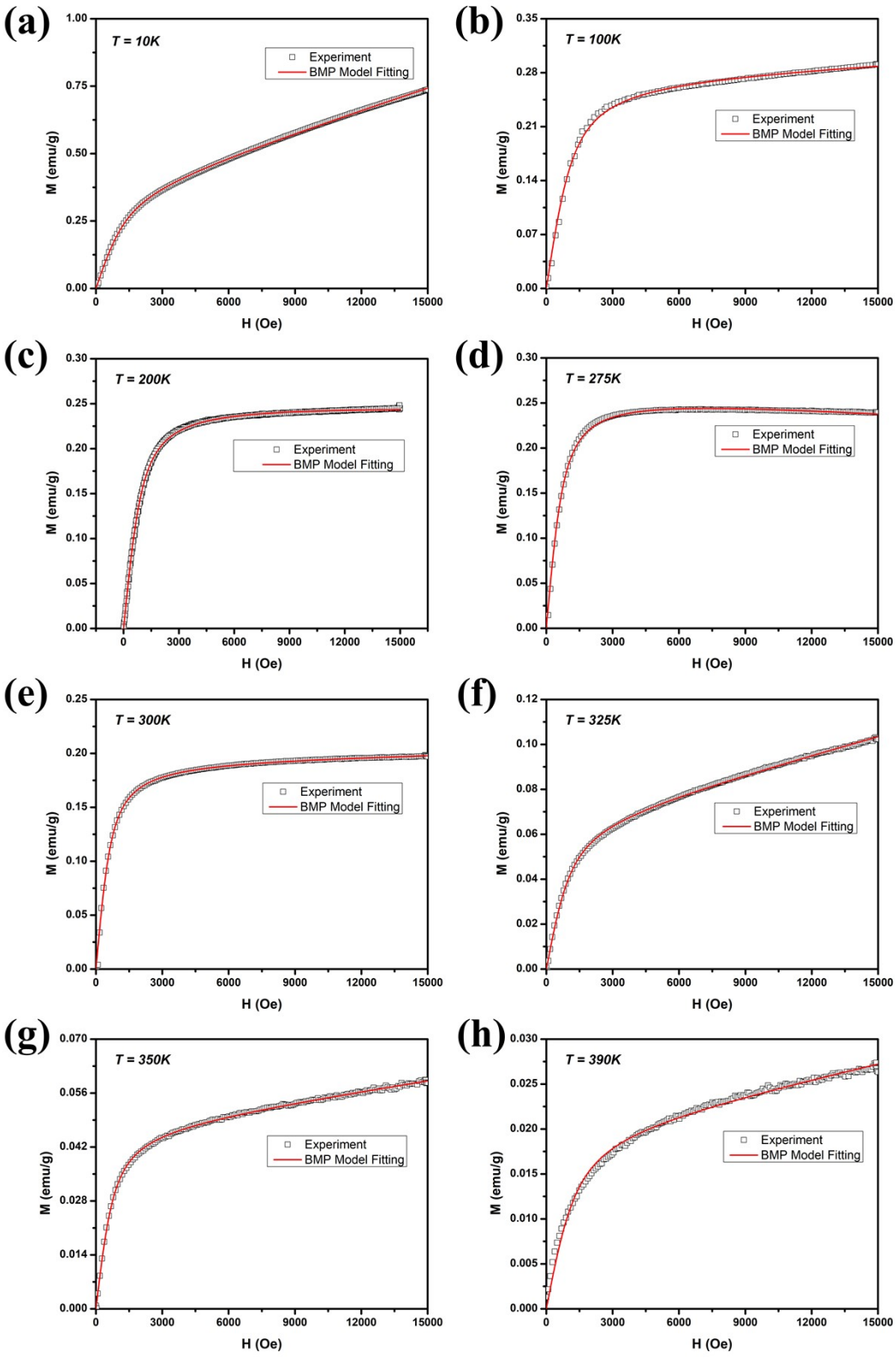


Figure S7. BMP model fitting of initial M-H relation for 5% Tb-Doped MoS_2 sample at different temperatures. (a) 10K. **(b)** 100K. **(c)** 200K. **(d)** 275K. **(e)** 300K. **(f)** 325K. **(g)** 350K. **(h)** 390K.

(a)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	5.40944E-6		
4	Adj. R-Square	0.99018		
5			Value	Standard Error
6	2% Tb MoS2 emu/g	A	0.05631	0.00149
7		B	0.00244	1.26906E-4
8		C	3.78536E-6	1.92833E-7

2% Tb MoS_2 BMP Fitting

(b)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	3.26389E-6		
4	Adj. R-Square	0.9992		
5			Value	Standard Error
6	5% Tb MoS2 emu/g	M0	0.21928	0.00143
7		Meff/kT	0.00279	3.98225E-5
8		Xm	3.878E-6	1.88849E-7

5% Tb MoS_2 BMP Fitting

(c)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	1.29883E-6		
4	Adj. R-Square	0.91922		
5			Value	Standard Error
6	Per mg	A	0.01319	3.91258E-4
7		B	0.00487	5.00092E-4
8		C	4.32665E-7	4.42352E-8

7% Tb MoS_2 BMP Fitting

(d)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	5.10721E-7		
4	Adj. R-Square	0.99887		
5			Value	Standard Error
6	8% Tb MoS2 emu/g	A	0.01353	5.22567E-4
7		B	0.00309	2.81077E-4
8		C	6.39153E-6	7.08626E-8

8% Tb MoS_2 BMP Fitting

Figure S8. Fitting statistics under the BMP model (Langevin relation) for Tb-doped MoS_2 samples at different doping concentrations. (a) 2% Tb^{3+} Doping. **(b)** 5% Tb^{3+} Doping **(c)** 7% Tb^{3+} Doping. **(d)** 8% Tb^{3+} Doping.

(a)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	9.10802E-6		
4	Adj. R-Square	0.99968		
5			Value	Standard Error
6	emug	A	0.35166	0.00118
7		B	0.0018	1.69719E-5
8		C	2.69409E-5	9.35488E-8

5% Tb MoS_2 BMP Fitting – 10K

(b)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	7.89629E-6		
4	Adj. R-Square	0.99744		
5			Value	Standard Error
6	emug	A	0.27671	0.00134
7		B	0.00201	2.94455E-5
8		C	1.38249E-6	1.08066E-7

5% Tb MoS_2 BMP Fitting – 100K

(c)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	1.40327E-6		
4	Adj. R-Square	0.99997		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.25541	1.59728E-4
7		Meff	0.00242	4.98951E-6
8		Xm	-3.33035E-7	1.36125E-8

5% Tb MoS_2 BMP Fitting – 200K

(d)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	3.56785E-6		
4	Adj. R-Square	0.99779		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.26763	5.61767E-4
7	5% Tb MoS2	Meff	0.00311	2.636E-5
8	5% Tb MoS2	Xm	-1.62259E-6	4.88298E-8

5% Tb MoS_2 BMP Fitting – 275K

(e)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	2.21457E-6		
4	Adj. R-Square	0.99795		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.19529	4.2268E-4
7	5% Tb MoS2	Meff	0.00348	3.26673E-5
8	5% Tb MoS2	Xm	4.14597E-7	3.73919E-8

5% Tb MoS_2 BMP Fitting – 300K

(f)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	4.05755E-7		
4	Adj. R-Square	0.9991		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.06512	2.21789E-4
7	5% Tb MoS2	Meff	0.00223	2.47635E-5
8	5% Tb MoS2	Xm	2.68423E-6	1.82744E-8

5% Tb MoS_2 BMP Fitting – 325K

(g)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	9.06939E-8		
4	Adj. R-Square	0.99917		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.04709	9.08959E-5
7	5% Tb MoS2	Meff	0.00301	2.30262E-5
8	5% Tb MoS2	Xm	8.729E-7	7.85797E-9

5% Tb MoS_2 BMP Fitting – 350K

(h)

	A	B	C	D
1	Model	Lang2 (User)		
2	Equation	A*((1/tanh(B*x))-1/(B*x))+(C*x)		
3	Reduced Chi-Sqr	1.81834E-7		
4	Adj. R-Square	0.99392		
5			Value	Standard Error
6	5% Tb MoS2	M0	0.0198	1.65902E-4
7	5% Tb MoS2	Meff	0.00183	4.37999E-5
8	5% Tb MoS2	Xm	5.43035E-7	1.31653E-8

5% Tb MoS_2 BMP Fitting – 390K

Figure S9. Fitting statistics under the BMP model (Langevin relation) for 5% Tb-doped MoS_2 sample at different temperatures. (a) 10K. **(b)** 100K. **(c)** 200K. **(d)** 275K. **(e)** 300K. **(f)** 325K. **(g)** 350K. **(h)** 390K.

(a)

	A	B	C	D
1	Model	Line		
2	Equation	y = A + B*x		
3	Reduced Chi-Sqr	7.1415E-37		
4	Adj. R-Square	0.99606		
5			Value	Standard Error
6	meff	A	9.2391E-17	1.10902E-18
7		B	4.485E-18	1.99186E-19

(b)

	A	B	C	D
1	Model	Pow2P2		
2	Equation	y = a*(1 + x)^b		
3	Reduced Chi-Sqr	3.52753E31		
4	Adj. R-Square	0.99943		
5			Value	Standard Error
6	N	a	1.49988E19	1.63671E18
7		b	-1.26834	0.04513

(c)

	A	B	C	D
1	Model	Cubic		
2	Equation	y = A + B*x + C*x^ 2 + D*x^3		
3	Reduced Chi-Sqr	1.47656E-36		
4	Adj. R-Square	0.99958		
5			Value	Standard Error
6	meff	A	-8.86054E-19	1.52124E-18
7		B	3.40676E-19	5.31311E-20
8		C	-1.01368E-21	4.30105E-22
9		D	4.94242E-24	9.21016E-25

Figure S10. Fitting statistics for parameter trends displayed under (a) Figure 8a. **(b)** Figure 8c. **(c)** Figure 8d.