Supplementary Information

Enhancing Magnetic Coupling in MN_4-Graphene via Strain Engineering

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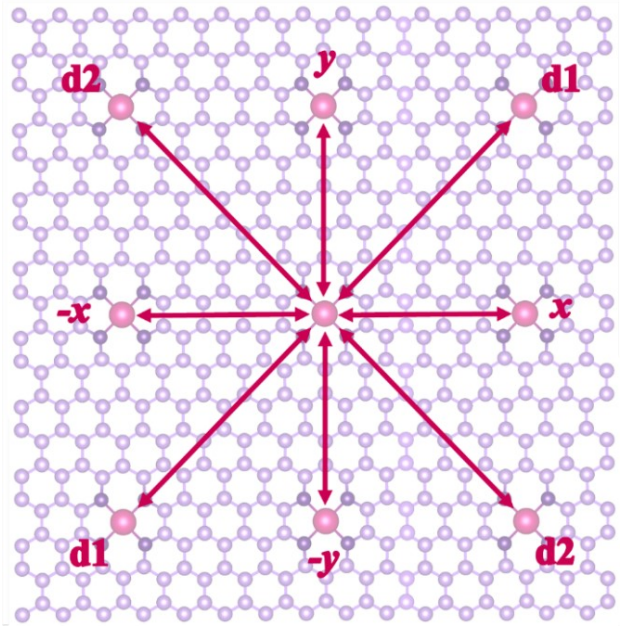

Fig. S1 The exchange couplings of the M atom located at the origin of a unit-cell with the neighboring atoms are shown with the purple double-head arrows along $\pm x$, $\pm y$, and the two main diameter (d1, d2) directions.

Table S1 The cohesive energy (CE) of the unstrained and strained MN_4-G layers. The CE defined as $CE = \frac{\left[E_{MN_4-G} - (N_C E_C + N_N E_N + N_M E_M)\right]}{q}$, where E_{MN_4-G} is the total energy of the studied layers, and E_C, E_N and E_M are the total energies of free C, N, and M atoms, respectively. N_C, N_N, and N_M are the number of C, N, and M atoms in the supercell, respectively. The parameter q stands for the total number of atoms in the supercell. For graphene, the CE per atom is -5.8275 eV.

Layer	CE/atom (eV)	Layer	CE/atom (eV)	Layer	CE/atom (eV)	Layer	CE/atom (eV)
Mn-Mn	-7.9709	Fe-Fe	-8.0017	Co-Co	-8.0198	Cu-Cu	-7.9752
Mn-Fe	-7.9698	Fe-Mn	-7.9976	Co-Mn	-8.0059	Cu-Mn	-7.9738
Mn-Co	-7.9658	Fe-Co	-8.0004	Co-Fe	-8.0043	Cu-Fe	-7.9750
Mn-Cu	-7.9730	Fe-Cu	-7.9980	Co-Cu	-8.0211	Cu-Co	-7.9725

Table S2 Fitting parameters of J_r in Eq. (8), for MnN_4-G layer under the strain. $k_F = [(k_F^x)^2 + (k_F^y)^2]^{1/2}$ is the Fermi wave number, k_F^x and k_F^y are components of the Fermi wave vector; and J_0 is a constant along $\pm x$, $\pm y$, and the two main diameter (d1, d2) directions.

MnN_4-G	Direction	$J_0(meV)$	$k_F^x(\text{Å}^{-1})$	$k_F^y(\text{Å}^{-1})$	$k_F(\text{Å}^{-1})$
$\eta = -0.36$	x	-5.548	0.118	------	------
	y	-0.003	------	0.002	------
	d1	-3.939	------	------	0.118
	d2	-3.949	------	------	0.118
$\eta = -0.20$	x	-6.313	0.121	------	------
	y	-0.007	------	0.002	------
	d1	-2.210	------	------	0.121
	d2	-0.900	------	------	0.121
$\eta = 0.00$	x	-75.610	0.123	------	------
	y	41.040	------	0.125	------
	d1	-4.402	------	------	0.175
	d2	-1.570	------	------	0.175
$\eta = 0.03$	x	-118.600	0.150	------	------
	y	-53.000	------	0.145	------
	d1	-15.560	------	------	0.209
	d2	-8.486	------	------	0.209

Table S3 Fitting parameters of J_r in Eq. (8), for FeN_4-G layer under the strain. Other parameters are detailed in the caption of Table S2.

FeN_4-G	Direction	$J_0(meV)$	$k_F^x(\text{Å}^{-1})$	$k_F^y(\text{Å}^{-1})$	$k_F(\text{Å}^{-1})$
$\eta = -0.16$	x	-0.600	0.117	------	------
	y	0.361	------	0.150	------
	d1	-0.100	------	------	0.190
	d2	0.200	------	------	0.190
$\eta = 0.00$	x	-0.164	0.125	------	------
	y	-0.029	------	0.022	------
	d1	-0.143	------	------	0.127
	d2	-1.300	------	------	0.127
$\eta = 0.20$	x	-1.46	0.108	------	------
	y	-0.084	------	0.037	------
	d1	0.188	------	------	0.114
	d2	-0.310	------	------	0.114
$\eta = 0.23$	x	-0.450	0.125	------	------
	y	0.753	------	0.091	------
	d1	0.191	------	------	0.155
	d2	0.319	------	------	0.155

Table S4 Fitting parameters of J_r in Eq. (8), for CoN_4-G layer under the strain. Other parameters are detailed in the caption of Table S2.

CoN_4-G	Direction	$J_0(meV)$	$k_F^x(\text{Å}^{-1})$	$k_F^y(\text{Å}^{-1})$	$k_F(\text{Å}^{-1})$
$\eta = 0.00$	x	-0.004	0.129	------	------
	y	-0.0004	------	0.029	------
	d1	-0.001	------	------	0.132
	d2	-0.001	------	------	0.132
$\eta = 0.16$	x	48.460	0.223	------	------
	y	0.099	------	0.097	------
	d1	0.240	------	------	0.243
	d2	-3.721	------	------	0.243
$\eta = 0.36$	x	0.833	0.099	------	------
	y	-0.100	------	0.046	------
	d1	0.109	------	------	0.109
	d2	-0.109	------	------	0.109
$\eta = 0.39$	x	19.65	0.221	------	------
	y	0.131	------	0.111	------
	d1	0.047	------	------	0.247
	d2	-1.391	------	------	0.247

Table S5 Calculated (k_F^{Cal}) and fitted (k_F^{Fit}) Fermi wave numbers for various strained and unstrained MN_4-G layers under the strain.

MnN_4-G			FeN_4-G			CoN_4-G		
η	$k_F^{Cal}(\text{Å}^{-1})$	$k_F^{Fit}(\text{Å}^{-1})$	η	$k_F^{Cal}(\text{Å}^{-1})$	$k_F^{Fit}(\text{Å}^{-1})$	η	$k_F^{Cal}(\text{Å}^{-1})$	$k_F^{Fit}(\text{Å}^{-1})$
-0.36	0.112	0.118	-0.16	0.106	0.190	0.00	0.105	0.132
-0.20	0.100	0.121	0.00	0.120	0.127	0.16	0.149	0.243
0.00	0.125	0.175	0.20	0.101	0.114	0.36	0.139	0.109
0.03	0.137	0.209	0.23	0.122	0.155	0.39	0.132	0.247