Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

1. Energetically favorable for hydrogenated carbon nanotubes

For each hydrogenated nanotube, we calculated the total energies under ferromagnetic (FM), antiferromagnetic (AFM) and nonmagnetic states, shown in Table S1. For clarity, the energy of ground state for each case is marked red. Apparently, the ground state is either FM or AFM, but not nonmagnetic. The difference between AFM and FM states is about 0~30 meV per unit cell (more than 20 atoms), except H10N1 (its FM state is much energetically lower than AFM one). Although FM is not the ground state for every configuration, it can be stabilized by an external magnetic field [Phys Rev Lett 102, 136810] [Nat Nanotech 3, 408] [Phys Rev B 88, 235434]. Thus, the FM state is realizable in practice, and the findings under this state are useful to devices' application.

Table S1 The total energies of each configuration under ferromagnetic, antiferromagnetic and nonmagnetic states. The ground state of each case is marked red.

Nanotube	Ferromagnetic (eV)	Antiferromagnetic (eV)	Nonmagnetic (eV)
H1N0	-3161.74395	-3161.71991	-3161.61318
H2N0	-3178.76726	-3178.77173	-3178.72014
H2N1	-3176.37012	-3176.39529	-3176.06173
H2N2	-3177.84870	-3177.85931	-3177.80382
H2N3	-3176.72490	-3176.71786	-3176.44193
H2N4	-3177.54127	-3177.54553	-3177.47368
H2N5	-3176.96837	-3176.93258	-3176.72436
H10N1	-3292.60867	-3292.08787	-3288.61535

Moreover, hydrogenation might be a good way to realize ferrimagnetic state in nanotubes, and no doubt the adsorption sites of hydrogen atoms will be crucial. However, possible hydrogenated configurations (or patterns) are quite a lot. It needs a systematic and detailed investigation. In the present work, we confine our studies to the FM state. As mentioned above, the FM state could be stabilized in practice, so the lack of studies on ferrimagnetic state does not affect the main conclusions.

2. The viability of ferromagnetic states in actual situation

We calculated the energies of each configuration under ferromagnetic and nonmagnetic states, as well as their difference, E_{non}-E_{FM}, shown in Table S2. As described above, the ground state of each tube is either ferromagnetic or antiferromagnetic, but not nonmagnetic. As shown in Table S2, the nonmagnetic state is at least 44.88 meV (per unit cell) energetically larger than the ferromagnetic one. For H10N1 tube, the energy difference even increases to about 4.0 eV, which is quite large. Besides the external magnetic field, anisotropy effect could also assist to preserve the spin ordering state at finite temperature [Phys Rev Lett 100, 047209] [Nat Nanotech 3, 408] [Phys Rev Lett 81, 208] [Phys Rev Lett 97, 216803]. So in actual situation, good viability of ferromagnetic state can be expected.

Table S2 The total energies of each configuration under ferromagnetic and nonmagnetic states, as well as their difference, i.e., the total energy of nonmagnetic state minus that of the ferromagnetic one.

Nanotube	Ferromagnetic (eV)	Nonmagnetic (eV)	E_{non}-E_{FM} (eV)

H1N0	-3161.74395	-3161.61318	0.13077
H2N0	-3178.76726	-3178.72014	0.04712
H2N1	-3176.37012	-3176.06173	0.30839
H2N2	-3177.84870	-3177.80382	0.04488
H2N3	-3176.72490	-3176.44193	0.28297
H2N4	-3177.54127	-3177.47368	0.06759
H2N5	-3176.96837	-3176.72436	0.24401
H10N1	-3292.60867	-3288.61535	3.99332

3. The unit cells of the linearly hydrogenated carbon nanotubes

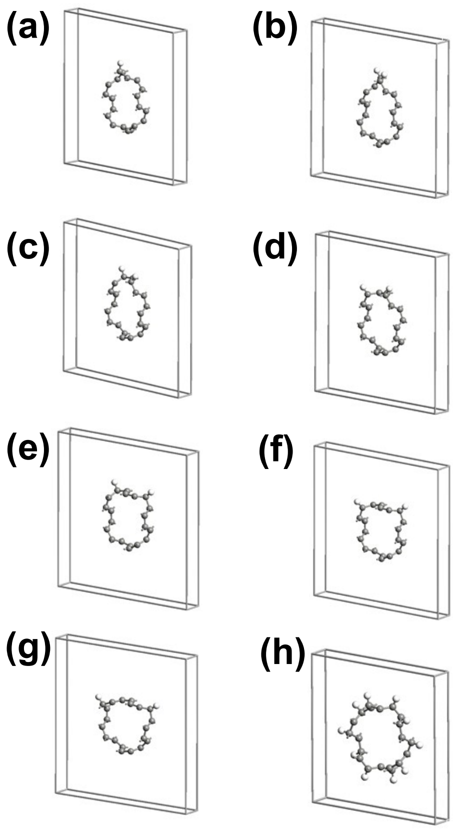

Figure S1 (a)-(h) The unit cells of the hydrogenated (5,5) tubes for H1N0, H2N0, H2N1, H2N2, H2N3, H2N4, H2N5 and H10N1, respectively. There is a one-to-one correspondence between Figure S1 and Figure 4.

4. The spin-Seebeck effect in (3,3) nanotube systems

As a demonstration to show the odd-even effect in nanotubes with other diameters, we calculated the spin-Seebeck current in (3,3) nanotubes by linear hydrogenation, shown in Figure S2. Considering the calculation cost, four configurations are investigated, i.e., H1N0, H2N0, H2N1 and H2N2. Apparently, the H2N0 and H2N2 cases exhibit spin-Seebeck diode effect, and the other two not. This is in accordance with the odd-even effect found in (5,5) tubes.

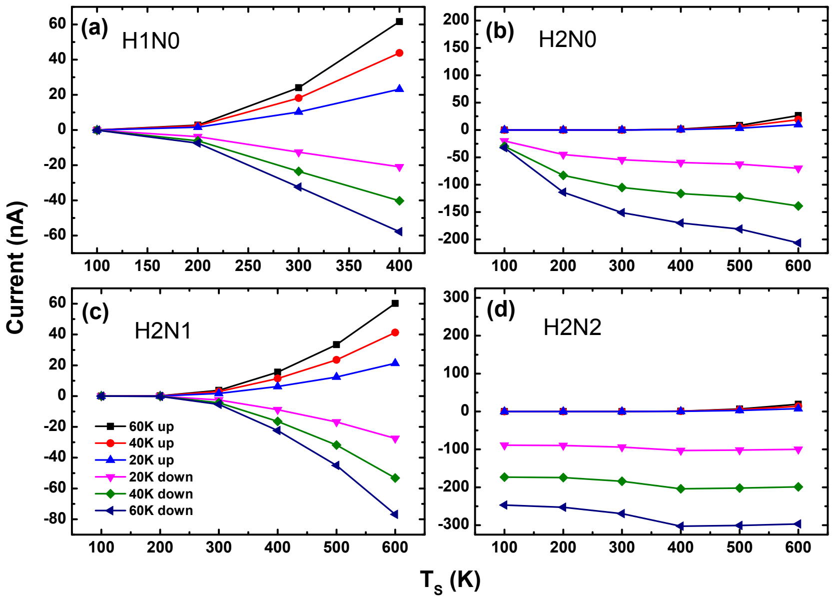

Figure S2 The spin dependent currents versus T_L for different T_{LR} ($T_{LR}=T_L-T_R=20$, 40 and 60 K) in linear hydrogenated (3,3) carbon nanotubes. The directions of the spin-up and spin-down currents are opposite, i.e., the spin-Seebeck effect. (a)-(d) Correspond to the systems of H1N0, H2N0, H2N1 and H2N2, respectively.