Supplementary Material

Electric field effect of sliding graphene / hexagonal boron nitride heterobilayer

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We investigated the periodic moiré patterns generated by rotating angles of the bilayers and obtained the relationship between the twisting angle and the periodic supercell size. The moiré pattern is a superstructure, and the periodicity is marked by rhombus in the Fig. 1. An equation of interlayer twisting angle θ as a function of integer *i* can be shown in the mathematical relationship: $\cos\theta = \frac{3i^2+3i+0.5}{3i^2+3i+1}$, where $i = 0, 1, 2$ ..., and the related superlattice vectors A_1, A_2 can be obtained by: $A_1 = ma_1 + na_2$, $A_2 = pa_1 + qa_2$ where a_1, a_2 are primitive unit-cell lattice vectors, and $m = i, n = i + 1, p = -(i + 1), q = (2i + 1)$. We set the Bravais lattice basis vector of the primitive cell $a_1 = (1/2, \sqrt{3}/2)a_0$, $a_2 = (-1/2, \sqrt{3}/2)a_0$ (a_0 is the lattice constant of graphene/h-BN). We rotate the upper layer of graphene so that there exist overlapping points (*m*, *n*) besides the origin point, then the supercell basis vector after rotation is $A_1 = ma_1 + na_2$, and let the angle rotated by graphene be $\theta/2$ we may obtain $cos^2(\frac{\theta}{2}) = \frac{(m^2+n^2-2mn)}{4(m^2+n^2+mn)}$.

Because of symmetry, if $\theta/2$ satisfies equation (1) below, then θ also satisfies that equation, *i.e.,* $cos\theta = 2cos^2(\frac{\theta}{2}) - 1 = -\frac{(m^2+n^2+4mn)}{2(m^2+n^2+mn)}$. Due to the symmetry of rotation, then $\theta = \theta + \pi$, so

$$cos\theta = \frac{(m^2+n^2+4mn)}{2(m^2+n^2+mn)} \quad (1)$$

where *m*, *n* are positive integers.

Let $n = i, m = i + 1, i = 0, 1, 2, 3$ ..., then we have

$$\cos\theta = \frac{3i^2+3i+0.5}{3i^2+3i+1} \quad (2)$$

Equation (2) is a special case of equation (1) and holds only when $m=n+1$ is satisfied.

Table S1 Structural parameters (θ, m, n, p, q) when twisting the graphene/h-BN heterobilayers. The total atom number is four times of the primitive cell number.

Angle	***m***	***n***	***p***	***q***	**Primitive cell No.**	**Atom No.**
60.000	0	1	-1	1	1	4
21.787	**1**	**2**	**-2**	**3**	**7**	**28**
13.174	**2**	**3**	**-3**	**5**	**19**	**76**
9.430	**3**	**4**	**-4**	**7**	**37**	**148**
7.341	4	5	-5	9	61	244
6.009	5	6	-6	11	91	364
5.086	6	7	-7	13	127	508
4.408	7	8	-8	15	169	676
3.890	8	9	-9	17	217	868
3.481	9	10	-10	19	271	1084
3.150	10	11	-11	21	331	1324
2.876	11	12	-12	23	397	1588
2.646	12	13	-13	25	469	1876
2.450	13	14	-14	27	547	2188
2.281	14	15	-15	29	631	2524
2.134	15	16	-16	31	721	2884
2.005	16	17	-17	33	817	3268
1.890	17	18	-18	35	919	3676
1.788	18	19	-19	37	1027	4108
1.696	19	20	-20	39	1141	4564
1.614	20	21	-21	41	1261	5044
1.539	21	22	-22	43	1387	5548
1.470	22	23	-23	45	1519	6076
1.408	23	24	-24	47	1657	6628
1.350	24	25	-25	49	1801	7204
1.297	25	26	-26	51	1951	7804
1.248	26	27	-27	53	2107	8428
1.203	27	28	-28	55	2269	9076
1.161	28	29	-29	57	2437	9748
1.121	29	30	-30	59	2611	10444
1.085	30	31	-31	61	2791	11164
1.050	31	32	-32	63	2977	11908
1.018	32	33	-33	65	3169	12676
0.987	33	34	-34	67	3367	13468
0.959	34	35	-35	69	3571	14284
0.932	35	36	-36	71	3781	15124
0.906	36	37	-37	73	3997	15988
0.882	37	38	-38	75	4219	16876
0.859	38	39	-39	77	4447	17788
0.837	39	40	-40	79	4681	18724
0.817	40	41	-41	81	4921	19684

Table S2 The summary of our calculated magnitude of the binding energy for the three stacking orders at different interlayer distances.

AB Stacking		AA Stacking		BA Stacking	
Distance (Å)	E_b (meV)	Distance (Å)	E_b (meV)	Distance (Å)	E_b (meV)
2.83	-27.231	3.04	-21.762	3	-24.041
2.88	-45.241	3.09	-37.132	3.05	-39.37
2.93	-59.771	3.14	-49.512	3.1	-51.76
2.98	-71.341	3.19	-59.332	3.15	-61.62
3.03	-80.391	3.24	-66.972	3.2	-69.33
3.08	-87.311	3.29	-72.782	3.25	-75.21
3.13	-92.421	3.34	-77.052	3.3	-79.56
3.18	-96.011	3.39	-80.042	3.35	-82.6
3.23	-98.331	3.44	-81.972	3.4	-84.56
3.28	-99.583	3.49	-83.002	3.45	-85.62
3.33	-99.951	3.54	-83.302	3.5	-85.942
3.38	-99.601	3.59	-82.998	3.55	-85.6
3.43	-98.661	3.64	-82.209	3.6	-84.77
3.48	-97.241	3.69	-81.017	3.65	-83.55
3.53	-95.441	3.74	-79.509	3.7	-82
3.58	-93.341	3.79	-77.759	3.75	-80.18
3.63	-91.001	3.84	-75.809	3.8	-78.18
3.68	-88.501	3.89	-73.719	3.85	-76.03
3.73	-85.871	3.94	-71.539	3.9	-73.827

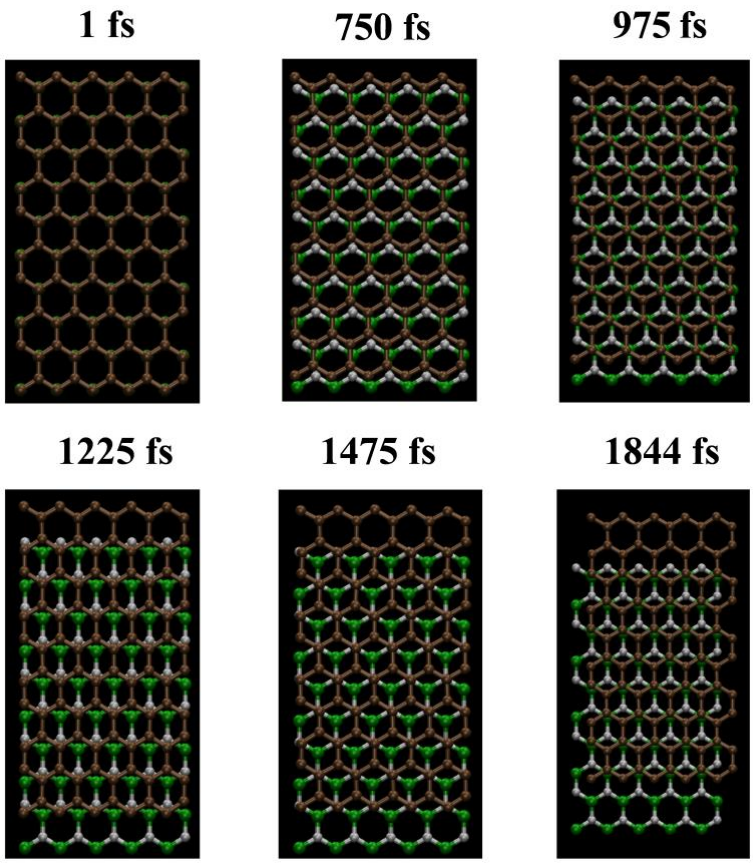

Fig. S1 The sliding motion snapshots of graphene/h-BN heterobilayer supercells using AIMD simulation under the action of external electric field.

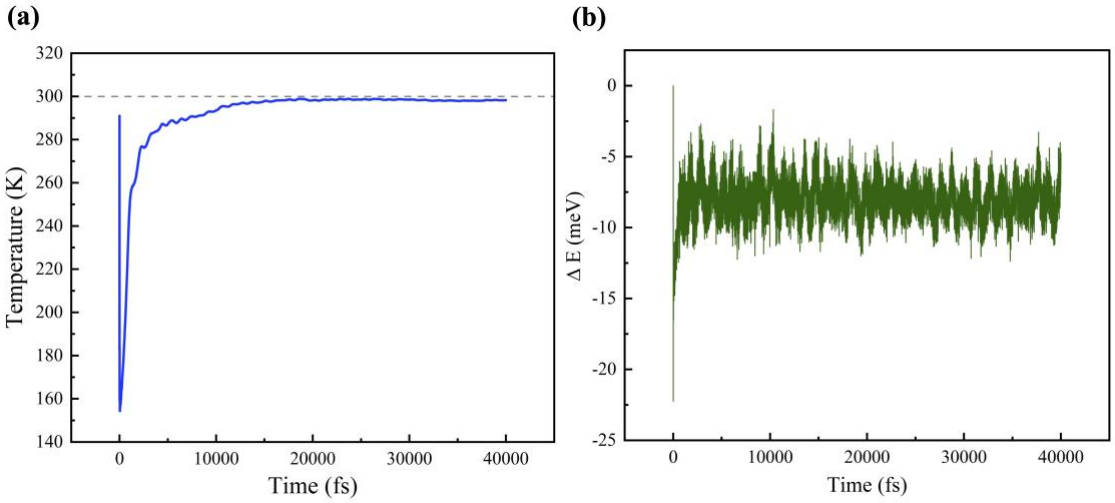

Fig. S2 (a) Temperature variation of the system within 40 ps from our AIMD simulation. (b) The evolution of total energy of the graphene/h-BN heterobilayer at the temperature of 300 K.

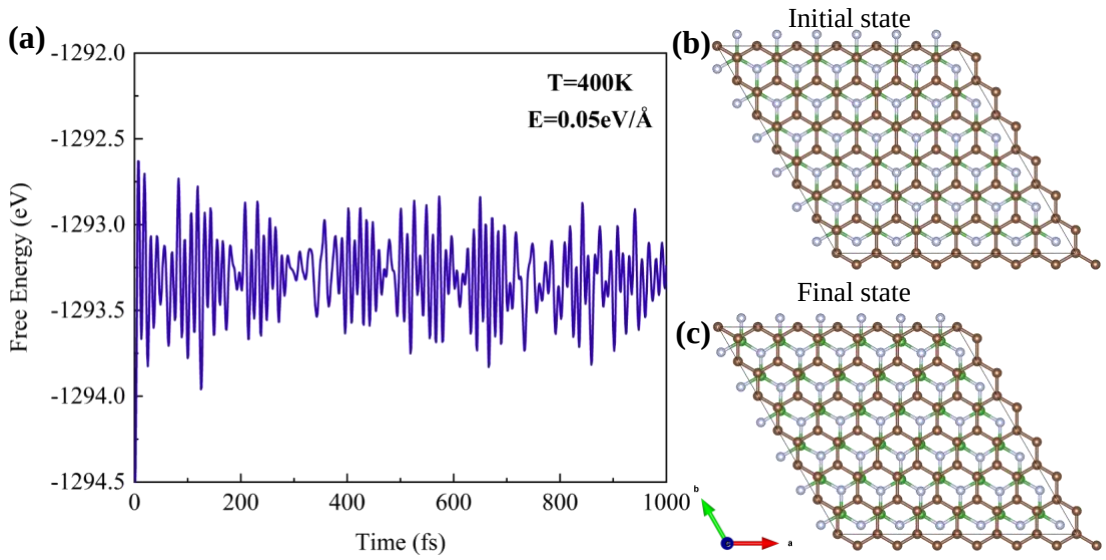

Fig. S3 (a) The time-dependent energy evolution of graphene/h-BN heterobilayer in AIMD simulations at 400 K with an electric field strength of 0.05 eV/Å. (b) and (c) are the initial and final state of the graphene/h-BN heterobilayer.