ELECTRONIC SUPPLEMENTARY INFORMATION

Open-shell jellium aromaticity in metal clusters

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Table S1 Cartesian coordinates of Be clusters under analysis.

Be_3^{+1}

1	4	0	0.000000	0.999684	-0.577173
2	4	0	0.000000	-0.999684	-0.577173
3	4	0	0.000000	0.000000	1.154346

Be_4

1	4	0	-0.414435	-0.585841	1.014686
2	4	0	-0.414435	-0.585841	-1.014686
3	4	0	-0.414435	1.171686	0.000000
4	4	0	1.243304	-0.000003	0.000000

Be_6^{-1}

1	4	0	0.000010	0.000020	-1.360165
2	4	0	1.486090	-0.442750	0.000010
3	4	0	-0.442630	-1.485978	-0.000002
4	4	0	0.000033	0.000038	1.360154
5	4	0	-1.486115	0.442481	0.000025
6	4	0	0.442612	1.486189	-0.000023

Be_7^{+1}

1	4	0	0.000000	0.000000	1.726180
2	4	0	0.000000	1.640908	0.533156
3	4	0	-1.246201	0.000000	0.000113
4	4	0	0.000000	-1.014402	-1.396358
5	4	0	1.246201	0.000000	0.000113
6	4	0	0.000000	-1.640908	0.533156
7	4	0	0.000000	1.014402	-1.396358

Be_9

1	4	0	-0.519649	-1.167715	-1.074817
2	4	0	1.272691	0.134515	-1.074850
3	4	0	0.811873	1.823038	0.001106
4	4	0	1.173433	-1.614379	-0.000506
5	4	0	1.271692	0.133115	1.075538
6	4	0	-0.752261	1.034142	1.074542
7	4	0	-0.751744	1.035735	-1.073938
8	4	0	-0.520534	-1.169552	1.073520
9	4	0	-1.985502	-0.208900	-0.000595

Be_9^{-1}

1	4	0	-0.370542	1.186649	1.094931
2	4	0	1.214724	-0.273021	1.094737
3	4	0	0.587173	-1.875258	-0.001406
4	4	0	1.332303	1.444985	0.000638
5	4	0	1.212731	-0.271654	-1.096054
6	4	0	-0.843280	-0.914415	-1.094744
7	4	0	-0.843053	-0.915746	1.094878
8	4	0	-0.372042	1.188789	-1.094056
9	4	0	-1.918014	0.429671	0.001076

Be_{10}^{+1}

1	4	0	-0.836055	1.557510	0.013801
2	4	0	0.836287	1.112694	-1.093142
3	4	0	2.222491	0.000136	0.000012
4	4	0	-0.835364	0.013805	-1.556671
5	4	0	0.836287	-1.093681	-1.112205
6	4	0	0.836300	-1.112762	1.093124
7	4	0	0.836287	1.093616	1.112189
8	4	0	-0.836009	-1.557586	-0.013808
9	4	0	-0.835337	-0.013811	1.556675
10	4	0	-2.224888	0.000079	0.000025

Be_{10}

1	4	0	-0.820758	-1.548350	-0.008785
2	4	0	0.821287	-1.102041	1.089930
3	4	0	2.165298	0.000011	0.000000
4	4	0	-0.820729	-0.008789	1.548167
5	4	0	0.821279	1.089924	1.102042
6	4	0	0.821278	1.102041	-1.089930
7	4	0	0.821285	-1.089925	-1.102045
8	4	0	-0.820768	1.548345	0.008786
9	4	0	-0.820731	0.008780	-1.548167
10	4	0	-2.167441	0.000002	0.000002

Be_{13}^{-1}

1	4	0	-0.587342	1.746895	-0.889200
2	4	0	-2.016049	0.335717	-0.117249
3	4	0	-0.759108	1.445194	1.233941
4	4	0	0.848740	0.157530	1.855134
5	4	0	-0.846143	-0.157032	-1.856668
6	4	0	0.761692	-1.444177	-1.233319
7	4	0	-1.183064	-1.636786	-0.331045
8	4	0	-1.129327	-0.646023	1.579637
9	4	0	1.129579	0.647209	-1.577915
10	4	0	1.183164	1.636127	0.329138
11	4	0	0.584765	-1.747577	0.889574
12	4	0	2.014933	-0.336289	0.117607
13	4	0	-0.001840	-0.000787	0.000367

Be_{14}^{+1}

1	4	0	1.209364	-0.409384	-1.696871
2	4	0	-1.066653	-0.401477	-1.528922
3	4	0	0.314092	1.558140	-1.363821
4	4	0	0.292322	1.998202	0.766586
5	4	0	0.292304	-1.998203	-0.766575
6	4	0	0.314099	-1.558112	1.363833
7	4	0	-1.449587	-1.514313	0.309690
8	4	0	-1.449590	1.514337	-0.309653
9	4	0	1.971560	-1.044508	0.235783

10	4	0	1.971567	1.044497	-0.235802
11	4	0	-1.066696	0.401446	1.528899
12	4	0	1.209412	0.409376	1.696854
13	4	0	0.148768	-0.000003	0.000014
14	4	0	-2.690960	0.000001	-0.000014

Table S2 Cartesian coordinates of Na clusters under analysis.

Na_5

1	11	0	0.000000	2.555188	-0.001732
2	11	0	-1.966068	0.000000	-1.130913
3	11	0	1.966068	0.000000	-1.130913
4	11	0	0.000000	0.000000	2.265290
5	11	0	0.000000	-2.555188	-0.001732

Na_8

1	11	0	0.719707	-2.341781	-0.054062
2	11	0	2.566329	-0.045721	1.854152
3	11	0	2.567405	0.045442	-1.854640
4	11	0	-0.717557	0.053548	-2.335042
5	11	0	0.719485	2.342278	0.054060
6	11	0	-0.716519	-0.054457	2.336149
7	11	0	-2.568883	1.853475	0.045044
8	11	0	-2.569966	-1.852784	-0.045660

Na_{13}

1	11	0	-1.563899	-2.530888	1.838796
2	11	0	1.563899	-0.966845	2.975206
3	11	0	-1.563899	0.966845	2.975206
4	11	0	-1.563901	3.128829	0.000000
5	11	0	1.563901	-3.128829	0.000000
6	11	0	1.563899	-0.966845	-2.975206
7	11	0	3.497124	0.000010	0.000000
8	11	0	1.563899	2.530888	1.838796
9	11	0	-1.563899	-2.530888	-1.838796
10	11	0	-3.497124	-0.000010	0.000000
11	11	0	1.563899	2.530888	-1.838796
12	11	0	-1.563899	0.966845	-2.975206
13	11	0	0.000000	0.000000	0.000000

Na_{18}

1	11	0	-0.935420	2.748646	2.061741
2	11	0	-0.935420	2.748646	-2.061741
3	11	0	-3.726241	1.901880	0.000000
4	11	0	-3.740337	-1.895150	0.000000
5	11	0	3.889522	-0.000695	0.000000
6	11	0	1.980593	-3.099800	0.000000
7	11	0	-0.145497	-0.004848	-4.278782
8	11	0	-3.246470	0.001099	-2.802667
9	11	0	-0.145497	-0.004848	4.278782
10	11	0	-3.246470	0.001099	2.802667
11	11	0	-0.941552	-2.747928	-2.045920
12	11	0	-0.941552	-2.747928	2.045920
13	11	0	-0.029568	0.006452	0.000000
14	11	0	2.545964	1.748978	3.020707
15	11	0	1.978935	3.103149	0.000000
16	11	0	2.546523	-1.753868	-3.021491

17	11	0	2.545964	1.748978	-3.020707
18	11	0	2.546523	-1.753868	3.021491

Na_{19}

1	11	0	2.036156	0.000000	-2.828304
2	11	0	-2.036156	0.000000	-2.828304
3	11	0	0.000000	2.826667	-3.239056
4	11	0	0.000000	4.756023	-0.003411
5	11	0	0.000000	-1.552217	-0.001798
6	11	0	0.000000	0.000000	3.490372
7	11	0	-3.293341	0.000000	1.089144
8	11	0	-3.076150	2.811058	-1.000632
9	11	0	3.293341	0.000000	1.089144
10	11	0	3.076150	2.811058	-1.000632
11	11	0	-1.897311	2.826460	2.619752
12	11	0	1.897311	2.826460	2.619752
13	11	0	0.000000	1.552217	-0.001798
14	11	0	3.076150	-2.811058	-1.000632
15	11	0	0.000000	-2.826667	-3.239056
16	11	0	0.000000	-4.756023	-0.003411
17	11	0	-1.897311	-2.826460	2.619752
18	11	0	-3.076150	-2.811058	-1.000632
19	11	0	1.897311	-2.826460	2.619752

Na_{20}

1	11	0	-2.308079	0.000000	2.433021
2	11	0	2.308079	0.000000	2.433021
3	11	0	0.000000	2.687656	2.898565
4	11	0	0.000000	4.718040	-0.354455
5	11	0	0.000000	-1.519349	-0.264030
6	11	0	0.000000	0.000000	-3.339374
7	11	0	3.430149	0.000000	-1.273844
8	11	0	3.108453	2.896417	0.650812
9	11	0	-3.430149	0.000000	-1.273844
10	11	0	-3.108453	2.896417	0.650812
11	11	0	2.010321	2.888398	-2.845261
12	11	0	-2.010321	2.888398	-2.845261
13	11	0	0.000000	1.519349	-0.264030
14	11	0	-3.108453	-2.896417	0.650812
15	11	0	0.000000	-2.687656	2.898565
16	11	0	0.000000	-4.718040	-0.354455
17	11	0	2.010321	-2.888398	-2.845261
18	11	0	3.108453	-2.896417	0.650812
19	11	0	-2.010321	-2.888398	-2.845261
20	11	0	0.000000	0.000000	5.238655

Table S3 Valence orbital energies (in a.u.) of Na and Be jellium clusters under analysis. Energy decreases from E_1 to E_7.

	Spin	**E_1**	**E_2**	**E_3**	**E_4**	**E_5**	**E_6**	**E_7**
Na_5	**3/2**	-0.112	-0.112	-0.128				
Na_{13}	**5/2**	-0.106	-0.106	-0.106	-0.106	-0.106		
Na_{19}	**1/2**	-0.102						
Be_3^{+1}	**3/2**	-0.391	-0.437	-0.437				
Be_6^{-1}	**5/2**	0.015	-0.009	-0.017	-0.017	-0.038		
Be_7^{+1}	**5/2**	-0.362	-0.362	-0.369	-0.392	-0.392		
Be_9^{-1}	**1/2**	-0.044						
Be_{10}^{+1}	**1/2**	-0.346						
Be_{13}^{-1}	**7/2**	-0.047	-0.047	-0.047	-0.047	-0.047	-0.047	-0.047
Be_{14}^{+1}	**7/2**	-0.319	-0.340	-0.343	-0.350	-0.356	-0.365	-0.372

Table S4 Full computational details.

Geometry optimizations without symmetry constraints and calculations of MCI and NICS(0) indices were performed with the Gaussian 09[34] and ESI-3D[35-37] packages of programs at the B3LYP/6-31G* level of theory.[38,39] Characterization of the stationary points was carried out by analytical frequency calculations. All presented geometries are minima. In all cases, we report the most stable minima found for each cluster. Although we tried different initial molecular structures for the optimization of each cluster, we can not fully guarantee that the minima found are the global minima. NICS(0) values have been computed through the gauge-including atomic orbital method (GIAO) implemented in Gaussian 09. The magnetic shielding tensor has been calculated for ghost atoms located at the atomic cluster centers determined by the non-weighted mean of the heavy atoms coordinates. Both $NICS(0)_{zz}$ and MCI have been calculated as the average of all three-member rings of the clusters. The atomization energy per atom has been calculated as the energy corresponding to these reactions: $[Na]_n \rightarrow nNa$ or $[Be]_n \rightarrow nBe$. In case or anionic or cationic clusters, the atomic energy of Be^- or Be^+ has been considered. On the other hand, the bond length alternation (BLA) is calculated as the difference between the longest and shortest bond length between two connected atoms in a cluster.

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Fig. S1 Atomization energies per atom (in eV) of the neutral Na_n clusters with n = 2 – 20 (in black). Sodium clusters that follow the open-shell jellium model in red; closed-shell clusters obeying the jellium model in green; the rest in black.