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RESEARCH ARTICLE

Graph attention network for global search of atomic clusters: A case study of Ag_n (n = 14–26) clusters

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Supporting Information

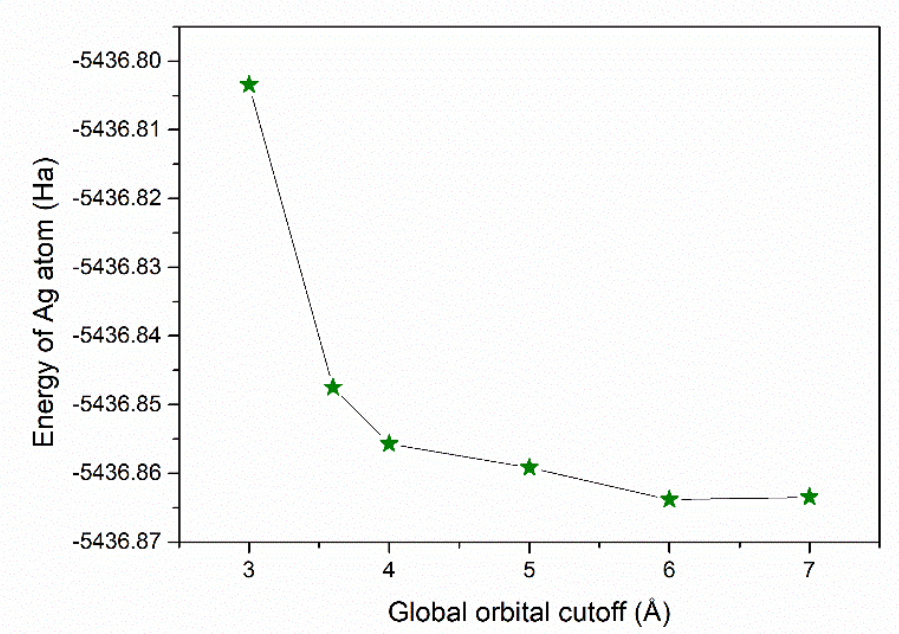


Fig. S1 Convergence test of global orbital cutoff for energy of Ag atom calculation by DMol[3].

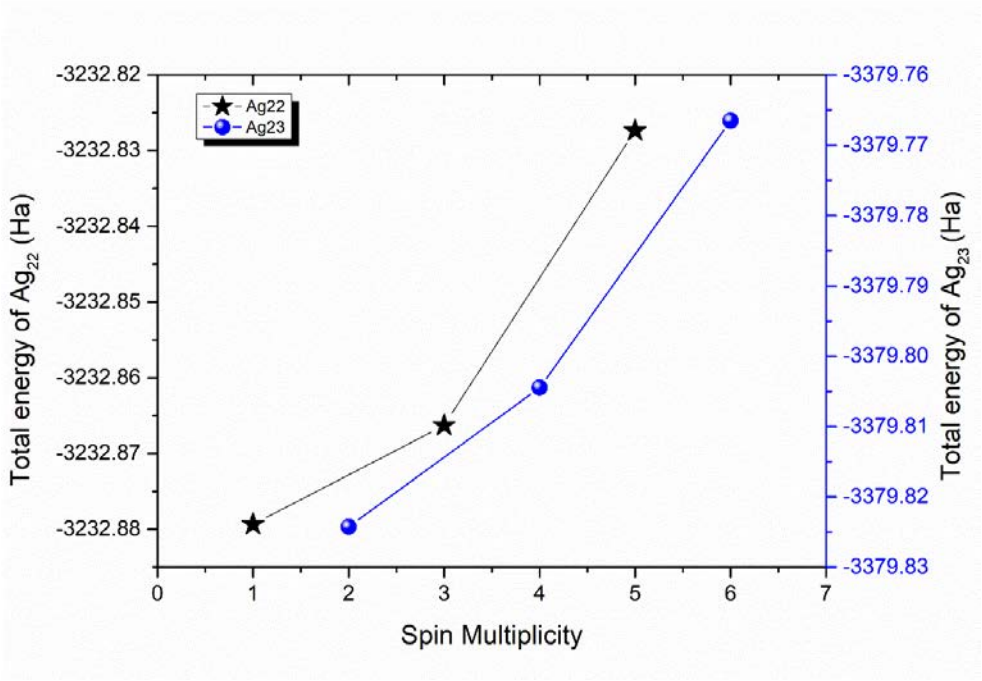
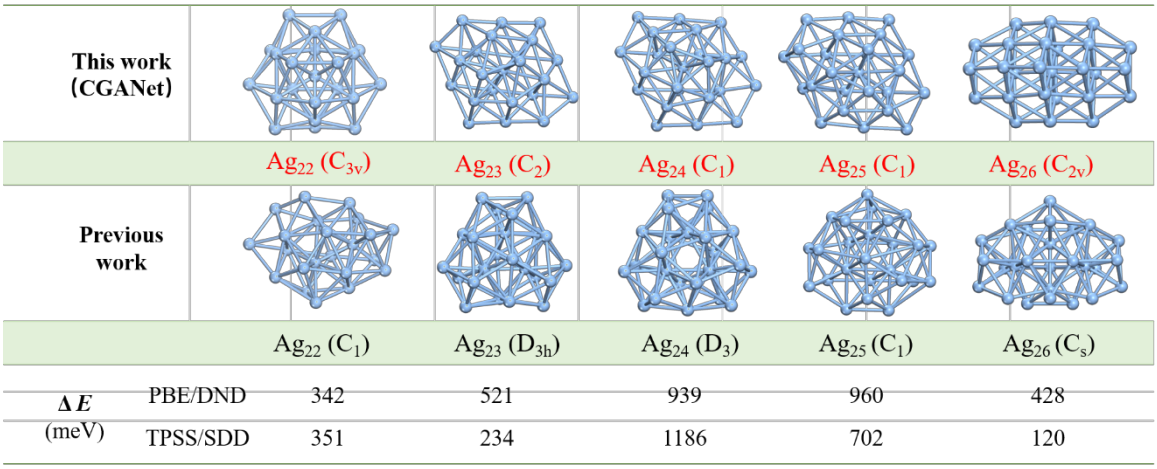


Fig. S2 The energies of Ag_{22} and Ag_{23} clusters calculated by Gaussian16 with different spin multiplicities. Ag_{22} reaches the energy minimum in spin doublet state, while Ag_{23} reaches the minimum in spin singlet state.



ΔE (meV)	PBE/DND	342	521	939	960	428
	TPSS/SDD	351	234	1186	702	120

Fig. S3 The newly discovered lowest-energy structures and symmetries of Ag_n (n = 22, 23, 24, 25, 26) clusters (red label) compared with the lowest-energy structures previously reported in Ref.1 (black label). Energy difference $\Delta E = E_{Ref} - E_{CGANet}$ are calculated by DMol^3 (PBE/DND) and Gaussian 16 (TPSS/SDD) in various functionals and basis.

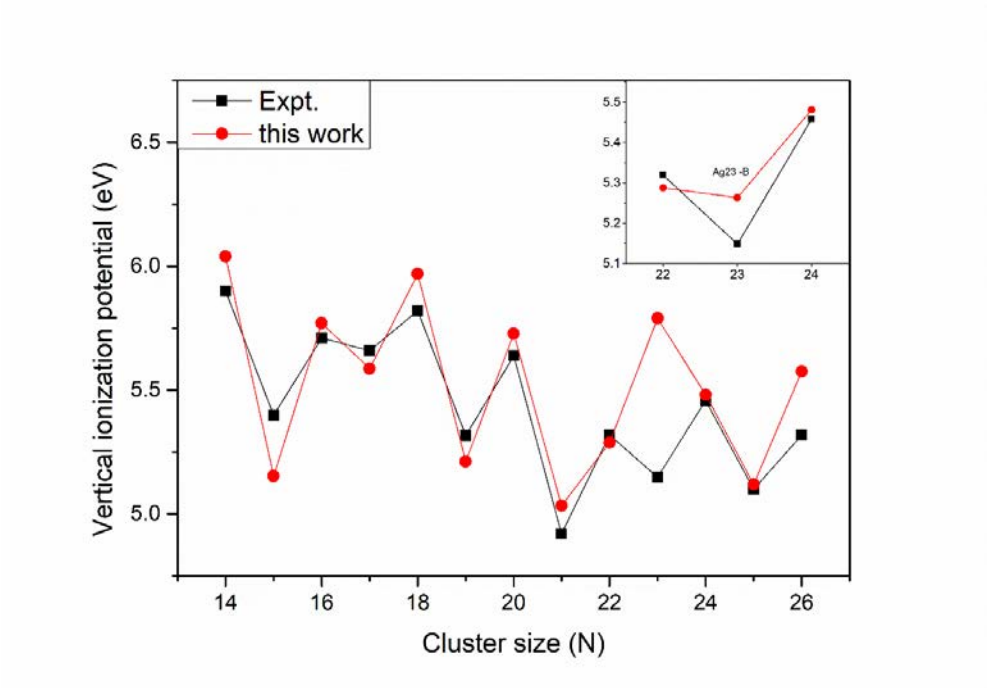


Fig. S4 Vertical ionization potentials for the lowest-energy structures of Ag_n (n = 14–26) clusters compared with experimental data.[2] The experimental data are well matched with the calculated data except for Ag_{23}. Illustration in the inset shows that the VIP of the isomer of Ag_{23}-B (see Fig. S5) is basically consistent with the experiment data.

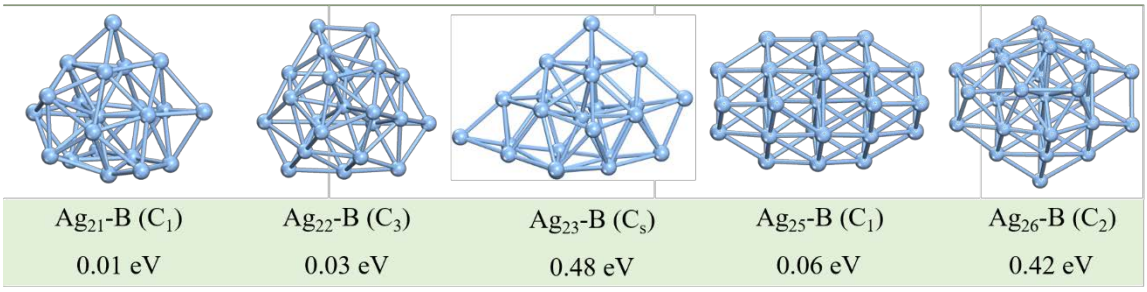


Fig. S5 Energy difference of new metastable structure for Ag_n (n = 21, 22, 23, 25, 26) clusters related to their ground-state structures calculated by DMol[3].

Table S1 Comparison of the experimental[3-5] and theoretical data calculated by DMol[3] and Gaussian 16 in various functionals and basis for Ag_2 dimer, including equilibrium bond length (r, in Å), binding energy (E_b, in eV), vertical ionization potential (VIP, in eV), and vibrational frequency (ω, in cm^{-1}).

Ag_2	**Expt.**	**DMol[3]**	**Gaussian**		
		PBE/DND	**PBE/SDD**	**HSE06/SDD**	**TPSS/SDD**
r (Å)	2.529	2.557	2.574	2.576	2.56
E_b (eV)	1.66	1.673	1.752	1.544	1.709
VIP (eV)	7.56	6.32	8.08	7.70	7.86
ω (cm^{-1})	192	193.6	186.1	186.6	192.7

Table S2 Average binding energy per atom of Ag_{16}, Ag_{20} and Ag_{24} dataset.

Ag cluster	Ag_{16}	Ag_{20}	Ag_{24}
***E*b (eV)**	1.627	1.704	1.726

Table S3 Cartesian coordinates (Å) of Ag_{22}, Ag_{23}, Ag_{24}, Ag_{25} and Ag_{26} clusters at TPSS/SDD level of theory.

	X	Y	Z
	-2.254	-2.2531	-2.2326
	-0.5424	-4.0642	-0.79
	-1.9783	-1.9781	0.5107
	-3.2489	2.5021	-0.7883
	-1.5735	0.4219	-1.4845
	0.4215	-1.5729	-1.4853
	-2.6509	0.7102	1.349
	-0.8251	3.0791	-2.2319
	-1.1682	-1.1687	3.1372
	0.7102	-2.6515	1.3481
Ag_{22}	0.0001	-0.0003	0.8976
	2.5014	-3.2488	-0.7899
	-0.7238	2.7019	0.5111
	1.1517	1.1522	-1.4845
	3.0783	-0.8247	-2.2331
	-0.427	1.5956	3.1374
	1.5964	-0.4285	3.1368
	2.7022	-0.7244	0.5102
	1.9412	1.9402	1.3484
	1.5626	3.7914	-0.7885
	3.7913	1.5625	-0.7893
	-4.0648	-0.5419	-0.7887

	X	Y	Z
Ag_{23}	4.925913	0.22944	-0.74261
	3.86125	-2.09873	0.324229
	-2.3196	0.604291	-1.44736
	-0.93884	-4.03707	-1.59931
	0.351164	1.363993	-1.91126
	-2.9676	-2.13815	-1.19878
	1.439492	-3.05526	-0.5451
	-0.35291	-1.36104	-1.91415
	-0.99767	-2.60711	0.767935
	-2.89971	-0.47843	1.086303
	2.318564	-0.60128	-1.44798
	0.001315	-0.00242	0.502023
	-3.86146	2.098736	0.327963
	0.989916	0.936357	3.002406
	1.527389	-1.8067	2.006398
	-1.44116	3.058534	-0.54312
	-0.98886	-0.94209	3.00189
	2.901348	0.477344	1.086532
	-1.52503	1.803975	2.005568
	0.996192	2.60417	0.769953
	-4.92603	-0.22859	-0.74028
	2.967536	2.140071	-1.19722
	0.938772	4.03996	-1.59403

	X	Y	Z
	0.833101	-2.61017	1.295184
	0.28873	0.006779	0.648097
	-2.7652	-2.68945	-1.85439
	-0.2649	1.414463	-1.82524
	-2.63921	0.049145	-1.34504
	2.131895	-2.68819	-1.21158
	2.166942	0.06826	-1.68824
	-0.25364	4.147933	-1.7384
	-2.52604	2.776163	-0.8102
	2.085505	2.779763	-1.0683
	-4.45871	-1.3955	0.087969
Ag_{24}	-0.36967	-3.91835	-0.92535
	-0.2532	-1.35164	-1.83394
	-4.72606	1.345133	0.028194
	4.556479	-1.32723	-1.49542
	4.347877	1.265365	-0.44057
	-0.16921	2.793779	0.6766
	-2.29375	0.826432	1.318756
	-1.79845	-1.96719	0.580659
	3.106858	-0.96794	0.798679
	-0.97867	-1.10668	3.049471
	2.337613	1.66387	1.494265
	-0.07695	1.489962	3.096482
	1.718642	-0.60472	3.162306

	X	Y	Z
	4.642962	0.931645	-0.11668
	2.589481	2.608177	-1.08557
Ag_{25}	2.468916	-0.1333	-1.43744
	0.36873	4.083828	-2.06569
	0.196253	1.374409	-1.95318
	4.731695	-1.68745	-1.09943

	0.028247	-1.40067	-1.79989
	2.224338	-2.89841	-0.96893
	3.130183	-1.19226	1.096421
	-2.46246	-2.50378	-2.25829
	-2.22764	0.166059	-1.25883
	-0.37716	-3.94304	-0.96941
	0.578016	-2.55417	1.304172
	-4.52182	-1.27091	-0.83573
	-2.0127	-2.05258	0.436331
	0.232355	0.088683	0.592986
	-2.063	2.937411	-1.19927
	2.47431	1.481024	1.516713
	0.247149	2.933569	0.465332
	-4.40978	1.54309	-0.32362
	1.429701	-0.64021	3.197944
	-2.10162	1.57806	1.251604
	-3.90135	-0.4125	1.709531
	-1.31341	-0.75103	2.80722
	0.048586	1.714337	2.993714

	X	Y	Z
Ag_{26}	2.266223	-1.78895	-2.82151
	-2.26723	-1.7891	-2.82206
	4.563092	-0.99329	-1.42166
	2.341048	0.660175	-1.49486
	0.000105	-0.19546	-2.86779
	4.567523	1.4488	-0.00243
	0.000107	2.236346	-1.45237
	-2.34115	0.661608	-1.49767
	2.303789	3.028664	-0.00378
	-0.00054	-0.18012	0.002329
	2.337056	-1.89888	0.001205
	4.562077	-0.98915	1.421735
	-0.0012	-2.61264	-1.43119

0.000102	-2.61091	1.437698
2.341653	0.665623	1.494864
2.266246	-1.78051	2.824837
-2.33649	-1.89672	0.002201
-2.26625	-1.78286	2.825619
-0.00041	-0.18801	2.869235
-4.56171	-0.98809	1.422899
0.000633	2.240997	1.449612
0.000538	4.602193	-0.00469
-2.34228	0.665962	1.495427
-2.30234	3.027653	-0.00364
-4.56757	1.449636	-0.00362
-4.56301	-0.99298	-1.42039

References

[1] Chen, Mingyang, et al. "Prediction of structures and atomization energies of small silver clusters, Ag_n, n<100. "*The Journal of Physical Chemistry A* 117.34 (2013): 8298-8313.

[2] Alameddin, George, et al. "Electronic and geometric structure in silver clusters." Chemical physics letters 192.1 (1992): 122-128.

[3] Morse, Michael D. "Clusters of transition-metal atoms. "*Chemical Reviews* 86.6 (1986): 1049-1109.

[4] Krämer, H-G., et al. "Sub-Doppler laser spectroscopy of silver dimers Ag_2 in a supersonic beam. "*Chemical physics letters*193.5 (1992): 331-334.

[5] Balasubramanian, K., and PingYi Feng. "The ionization potentials of Ag_n and Au_n and binding energies of Ag_n, Au_n, Ag_n^+ and Au_n^+ (n= 1–4)." *Chemical physics letters* 159.5-6 (1989): 452-458.