Supporting Information for the paper:

Manifestation of Geometric and Electronic Shell Structures of Metal Clusters in Intercluster Reactions

K. R. Krishnadas, Ananya Baksi, Atanu Ghosh, Ganapati Natarajan and Thalappil Pradeep*

Department of Chemistry, DST Unit of Nanoscience (DST UNS) and Thematic Unit of Excellence (TUE)
Indian Institute of Technology Madras
Chennai 600 036, India
E-mail: pradeep@iitm.ac.in

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Instrumentation

UV/Vis absorption spectroscopy: The UV/Vis spectra were recorded using a Perkin Elmer Lambda 25 UV/Vis spectrometer. Absorption spectra were typically measured in the range of 200-1100 nm.

Mass spectrometric measurements: Waters Synapt G2-Si High Definition Mass Spectrometer equipped with Electrospray ionization, matrix assisted laser desorption ionization and ion mobility separation was used. All the samples were analyzed in negative ion mode in electrospray ionization. The instrumental parameters were first optimized for $Ag_{44}(FTP)_{30}$ and other samples were analyzed using the similar setting with slight modification depending on the sample. About 0.1 mg of as prepared samples were diluted dichloromethane (DCM) and directly infused to the system without any further purification. The instrument was calibrated using NaI as calibrant. To get a well resolved spectrum of $[Ag_{44}(FTP)_{30}]^{4-}$, the instrumental parameters were as follows:

Sample concentration: 10 µg/mL
Solvent: DCM
Flow rate: 10-20 µL/min
Capillary voltage: 500-1000 V
Cone voltage: 0-20 V
Source offset: 0-10 V
Desolvation gas flow: 400 L/h
Trap gas flow: 2 mL/min
Capillary temperature: 60 ºC
Desolvation temperature: 80 ºC

All the mass spectra presented in the main paper and the Supporting Information are measured in a broad mass range, m/z 1000-20000. The selected regions of each of the broad range spectra showing the $[Au_xAg_{44-x}(FTP)_{30}]^{4-}$ clusters are presented in the main paper. The selected regions of each of the broad range spectra, showing the $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$ are presented in the Supporting Information.

Supporting Information 1

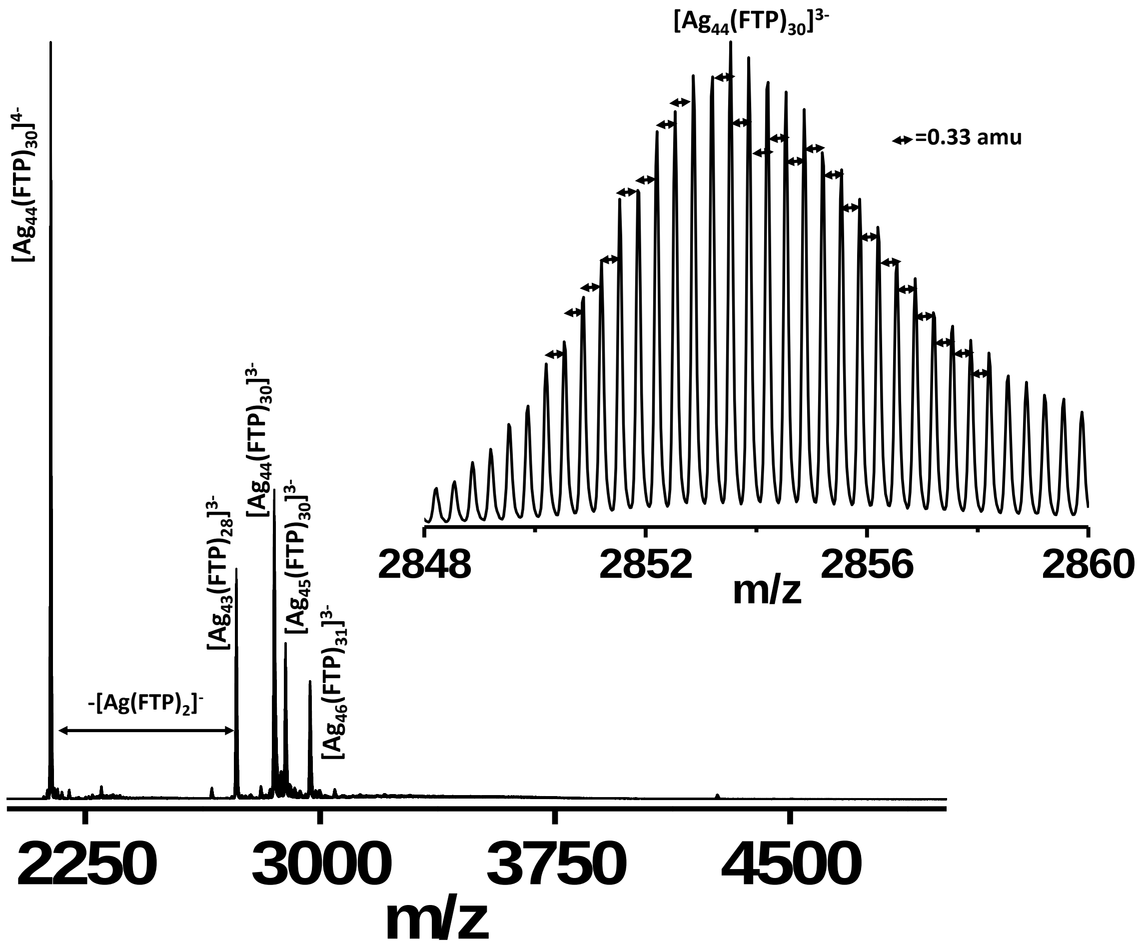

Figure S1. ESI MS spectrum of $Ag_{44}(FTP)_{30}$ showing its 4^- and 3^- charge states. Characteristic fragmentation of $[Ag_{44}(FTP)_{30}]^{4-}$ through the loss of $[Ag(FTP)_2]^-$ is also marked. Inset shows the experimental isotope pattern of $[Ag_{44}(FTP)_{30}]^{3-}$ which consists of a series of peaks with a separation of 0.33 amu, confirming the 3^- charge state.

Supporting Information 2

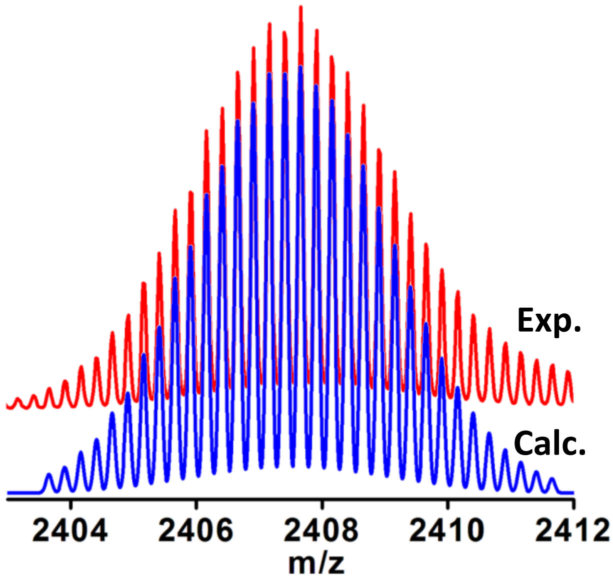

Figure S2. Theoretical (blue/lower) and experimental (red/upper) isotope distribution of $[Au_{12}Ag_{32}(FTP)_{30}]^{4-}$.

Supporting Information 3

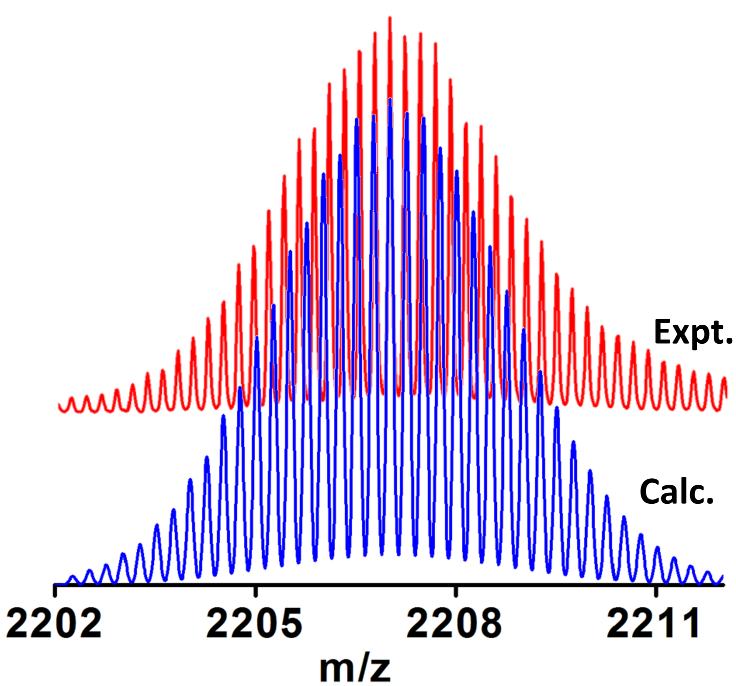

Figure S3. Theoretical (blue/lower) and experimental (red/upper) isotope distribution of $[Au_{3}Ag_{41}(FTP)_{30}]^{4-}$.

Supporting Information 4

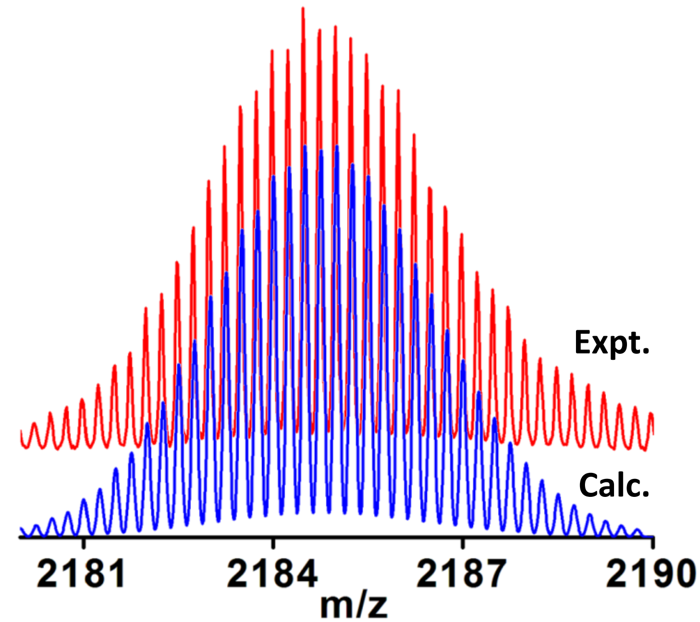

Figure S4. Theoretical (blue/lower) and experimental (red/upper) isotope distribution of $[Au_2Ag_{42}(FTP)_{30}]^{4-}$.

Supporting Information 5

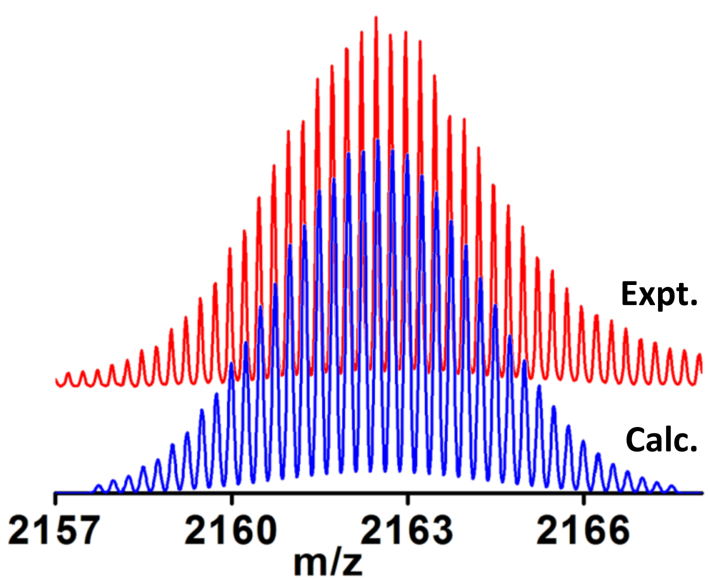

Figure S5. Theoretical (blue/lower) and experimental (red/upper) isotope distribution of $[Au_1Ag_{43}(FTP)_{30}]^{4-}$.

Supporting Information 6

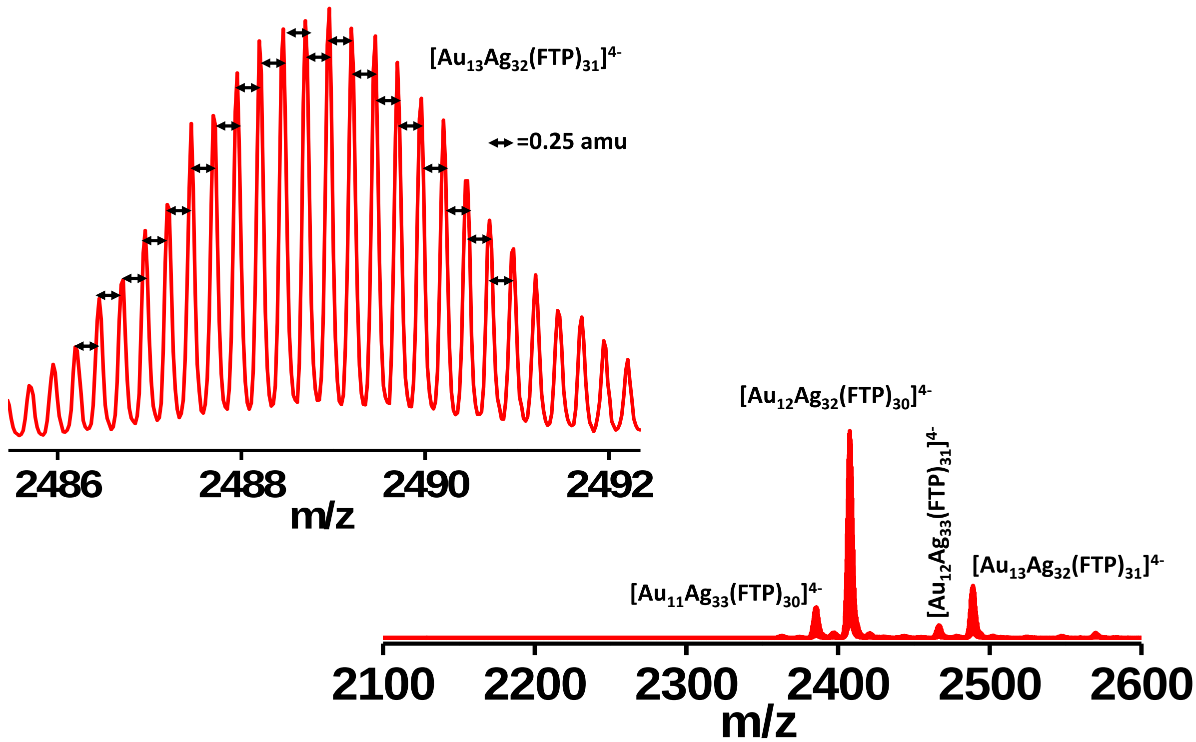

Figure S6. The mass spectra showing the assignment of the peaks labeled as '*' and '**' in Figure 1C. These peaks are due to clusters containing additional Ag-FTP and Au-FTP fragments and therefore do not belong to the general formula, $[Au_xAg_{44-x}(FTP)_{30}]^{4-}$. An expansion of the peak due to $[Au_{13}Ag_{32}(FTP)_{30}]^{4-}$ is given in the inset which confirms the isotopic patterns with peaks separated by 0.25 amu which confirms the charge state of this cluster.

Supporting Information 7

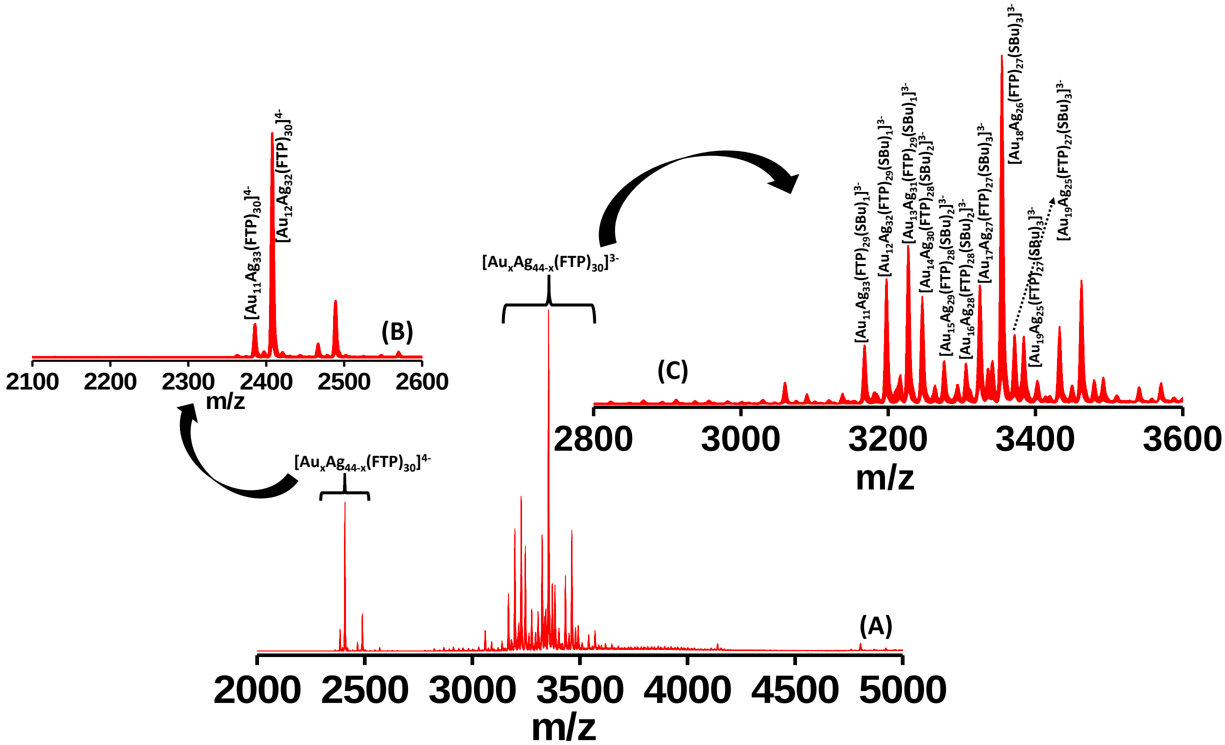

Figure S7. (A) A full range mass spectrum of a mixture of $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at a $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 2.8:1.0, selected portions of this spectra showing (B) the $[Au_xAg_{44-x}(FTP)_{30}]^{4-}$ clusters and (C) and $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$. The spectrum shown in (B) is the same as that shown in Figure 1C (red trace) in the main manuscript. The spectrum shown in (C) is the same spectra shown in Figure S16.

Supporting Information 8

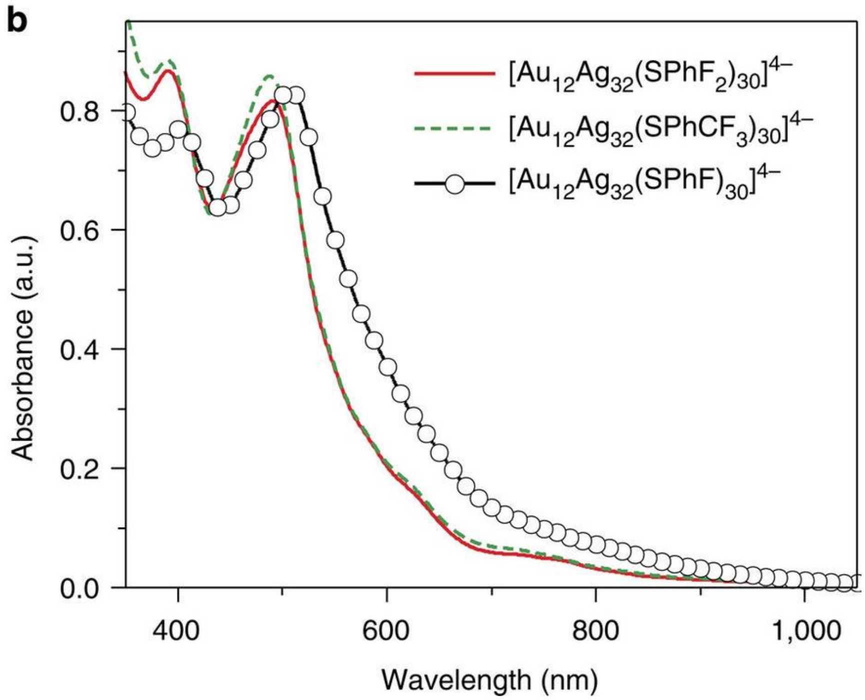

Figure S8. UV/Vis absorption spectra of $Au_{12}Ag_{32}(SR)_{30}$ clusters protected with various thiols. This figure is adapted from the paper, *Nat. Commun.* 2013, 2, 2422 by Zheng, N. *et al*. The absorption spectrum shown as black circles is similar to the spectrum shown in bottom trace corresponding to *t*=1 h, in Figure 1D.

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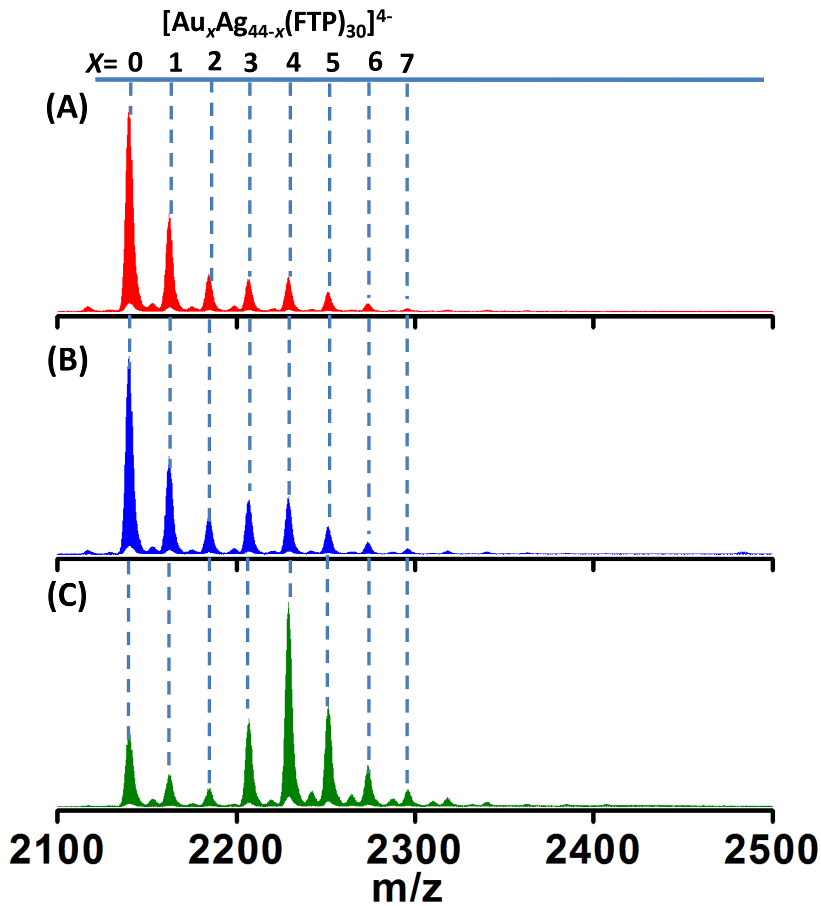

Figure S9. Time-dependent mass spectra measured within 2 min (A), after 10 min (B) and after 1 h (C) during the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at a $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 35.0:1.0.

Supporting Information 10

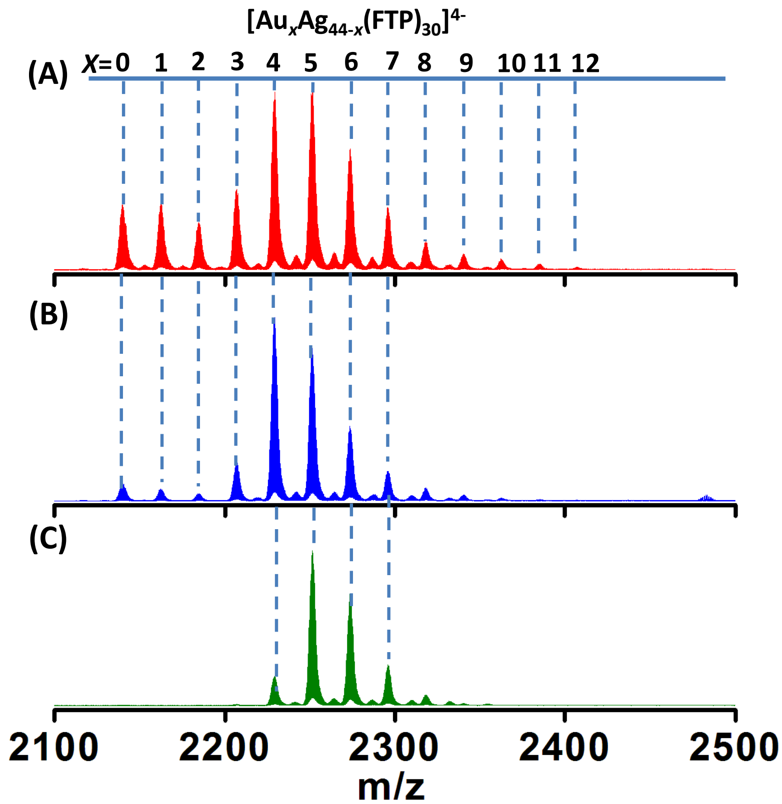

Figure S10. Time-dependent mass spectra measured within 2 min (A), after 10 min (B) and 1 h (C) during the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 14.0:1.0.

Supporting Information 11

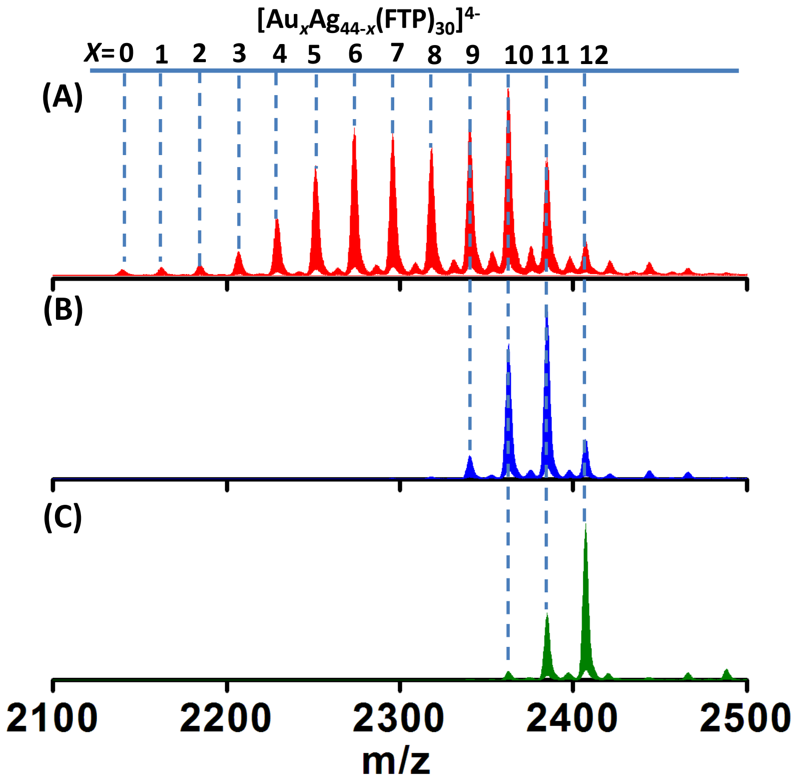

Figure S11. Time-dependent mass spectra measured within 2 min (A), after 10 min (B) and 1 h (C) during the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 5.0:1.0.

Supporting Information 12

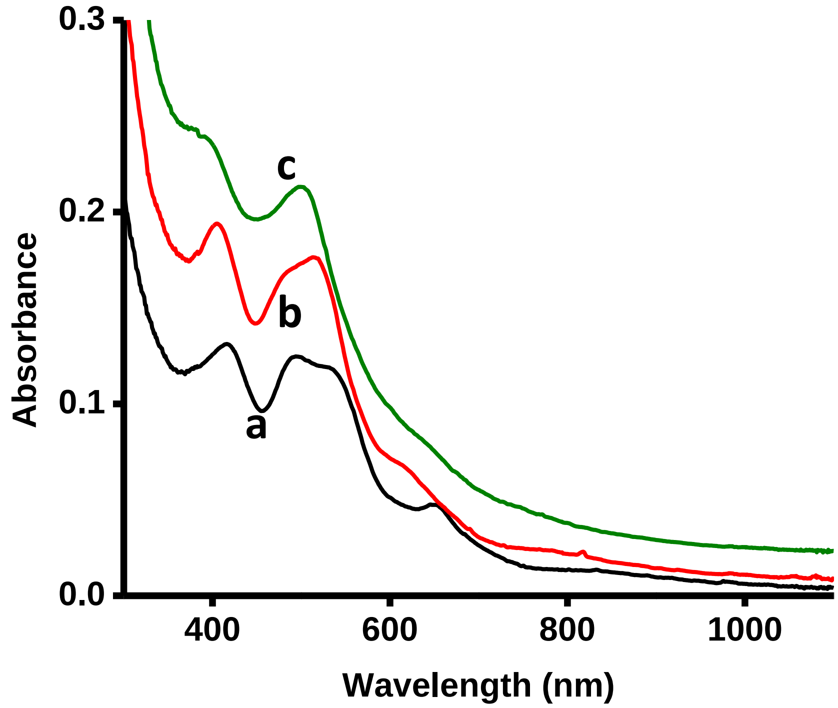

Figure S12. UV/Vis spectra of reaction mixtures, measured 1 h after mixing, at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratios of 35.0:1.0 (trace a), 14.0:1.0 (trace b) and 5.0:1.0 (trace c).

Supporting Information 13

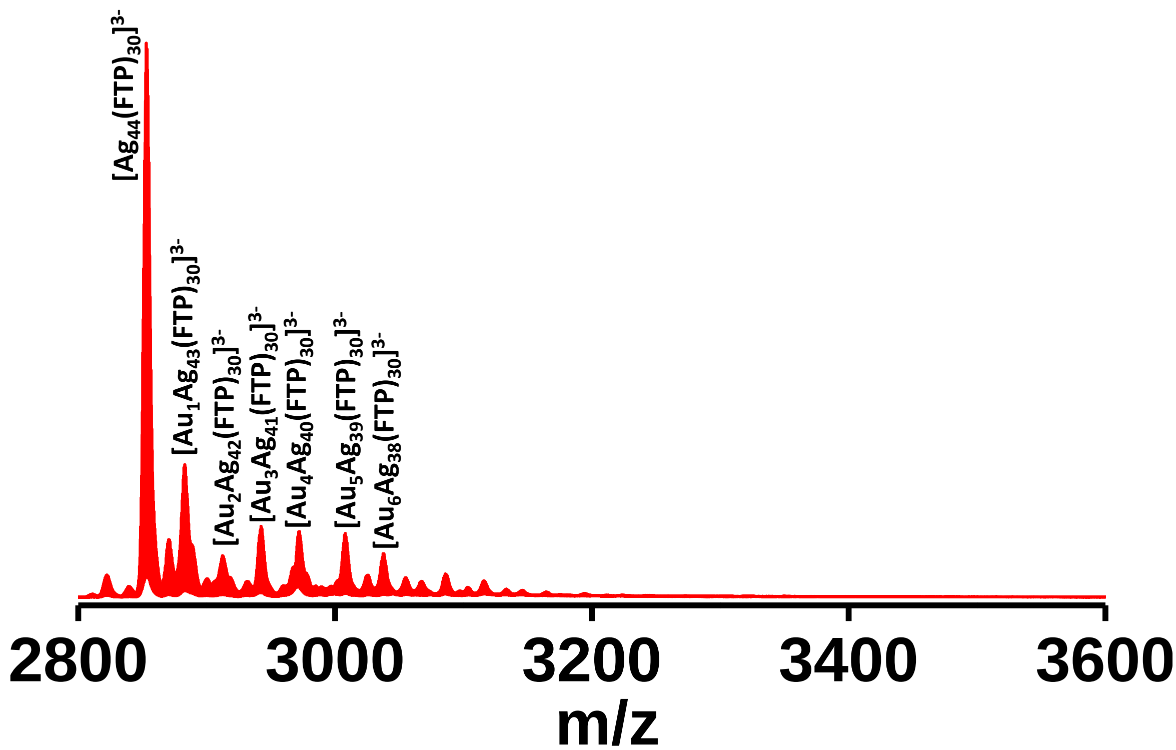

Figure S13. ESI MS spectra of $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$ formed from the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 35.0:1.0.

Supporting Information 14

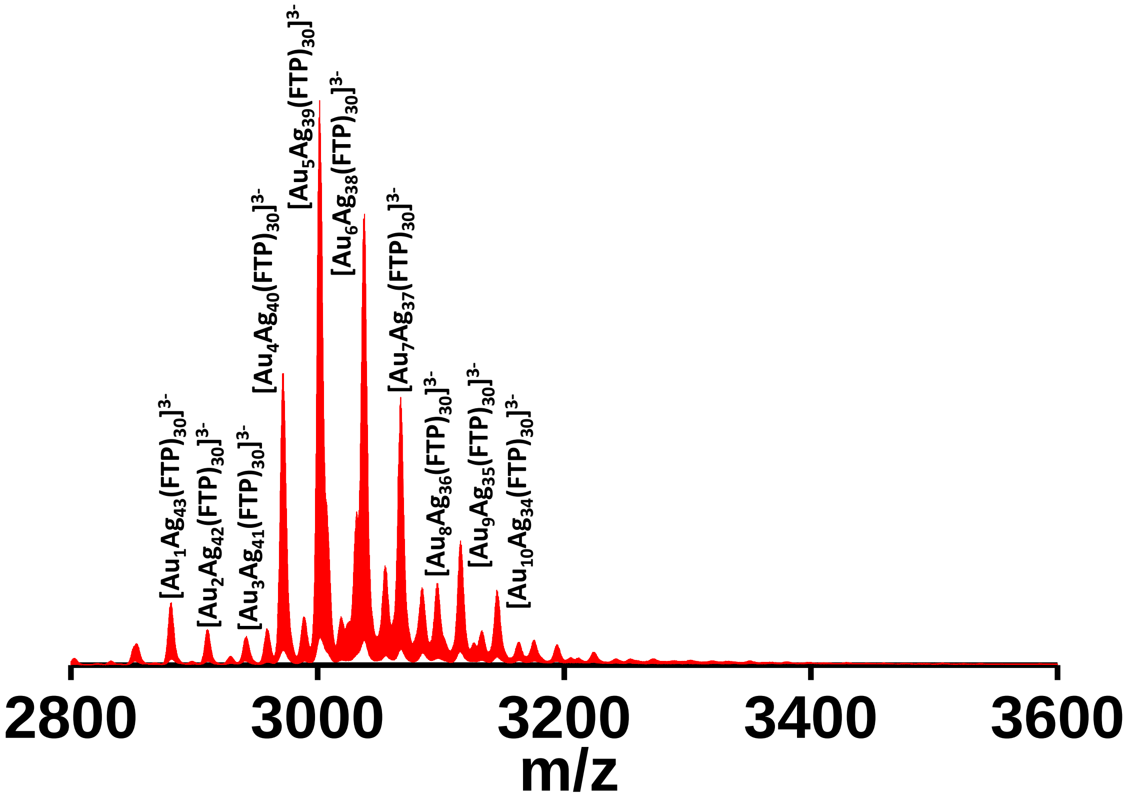

Figure S14. ESI MS spectra of $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$ formed from the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 14.0:1.0.

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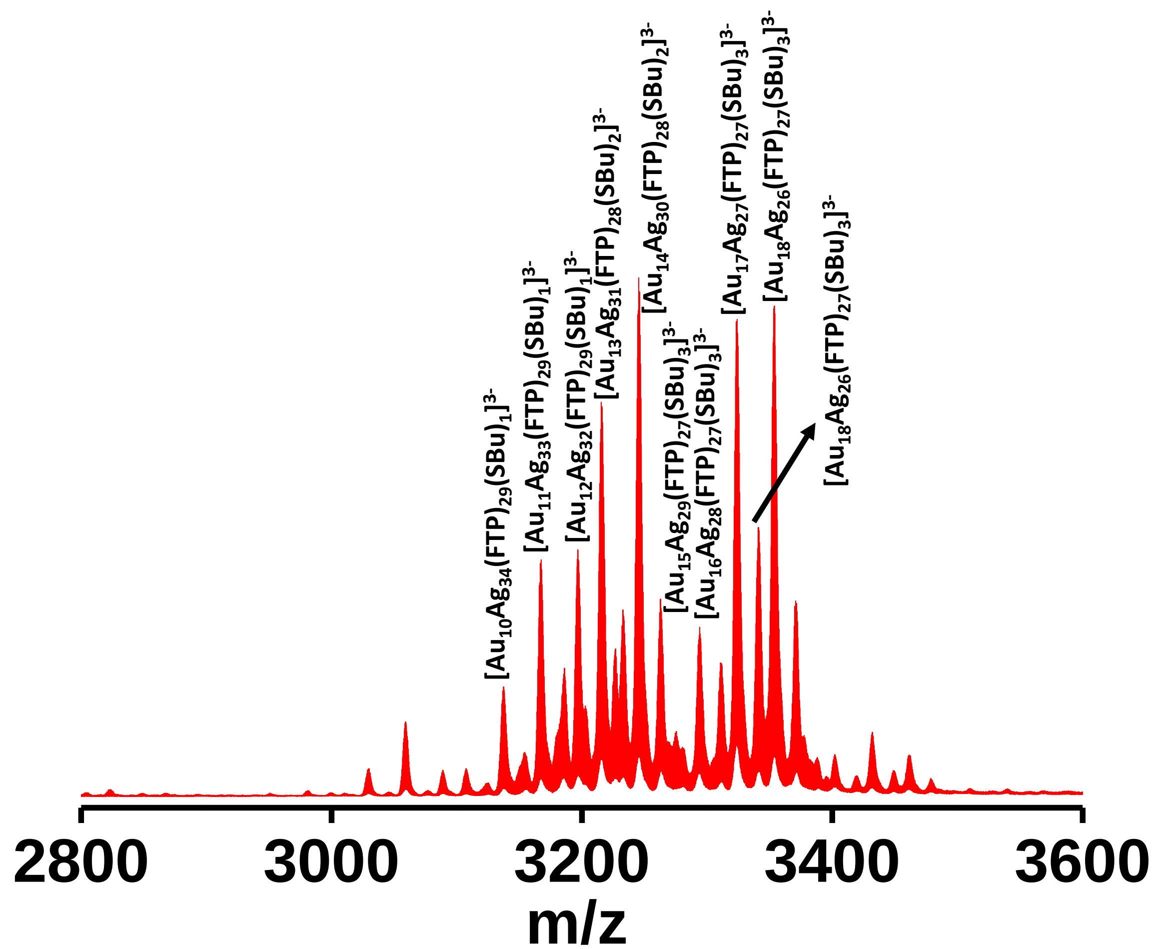

Figure S15. ESI MS spectra of $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$ formed from the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 5.0:1.0.

Supporting Information 16

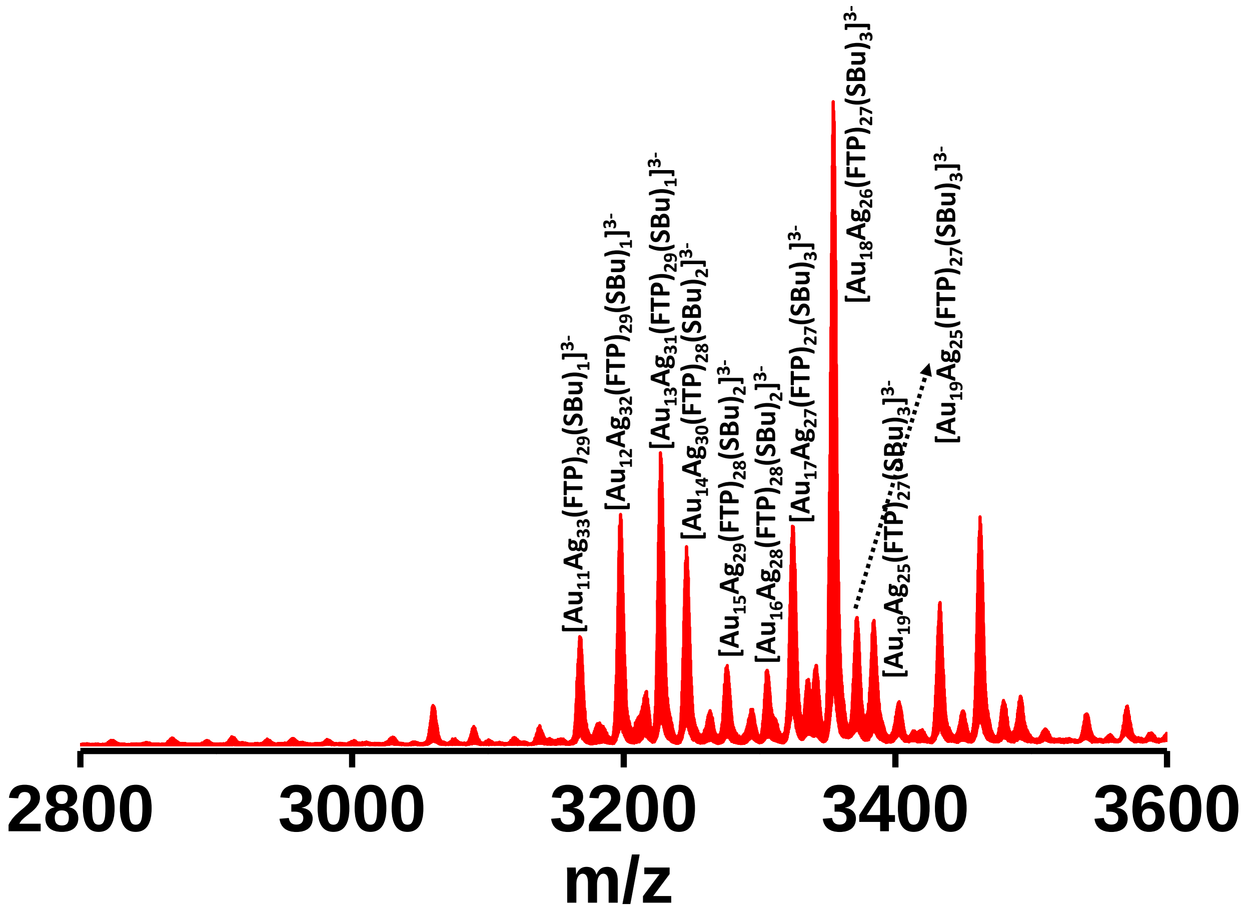

Figure S16. ESI MS spectra of $[Au_xAg_{44-x}(FTP)_{30}]^{3-}$ formed from the reaction between $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratio of 2.8:1.0.

Supporting Figure S17

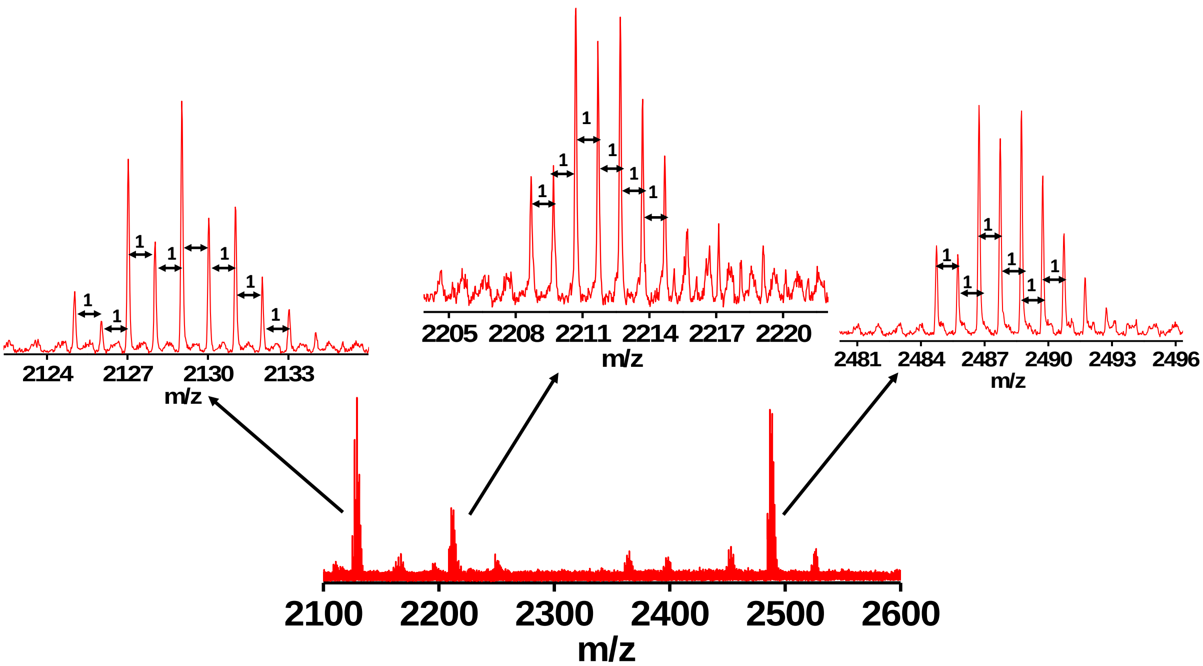

Figure S17 Expansion of the peaks in Figure 3C showing the unit charge state of the species formed. The prominent peaks in the mass spectra are expanded to show the unit separation between the peaks indicating that these species have unit negative charge. This shows that these peaks are not due to any $Au_xAg_{44-x}(FTP)_{18}$ species.

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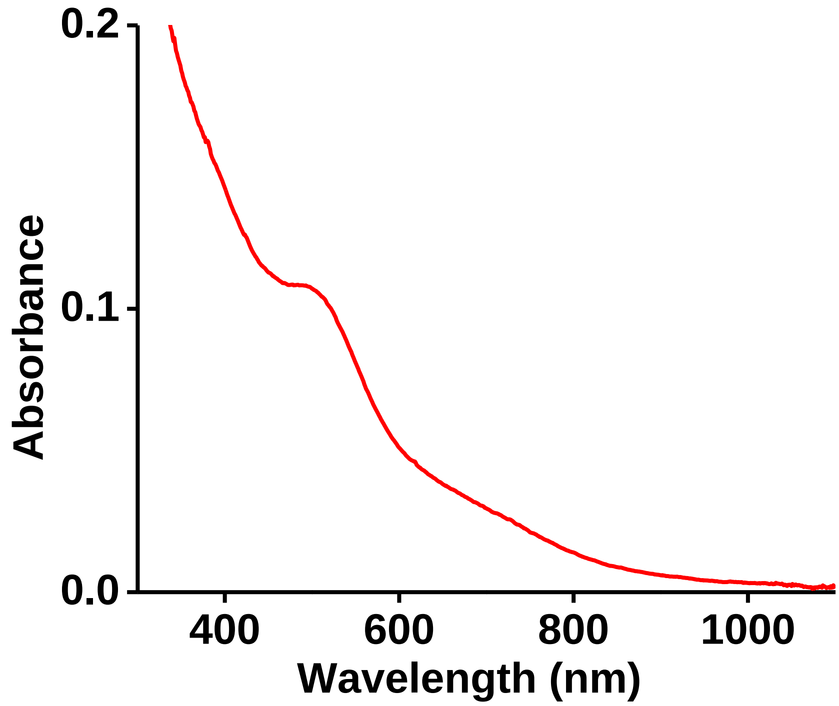

Figure S18. UV/Vis spectra of a mixture of $Ag_{44}(FTP)_{30}$ and $Au_{25}(FTP)_{18}$ at $Ag_{44}(FTP)_{30}$:$Au_{25}(FTP)_{18}$ molar ratios of 0.9:1.0.

Supporting Information 19

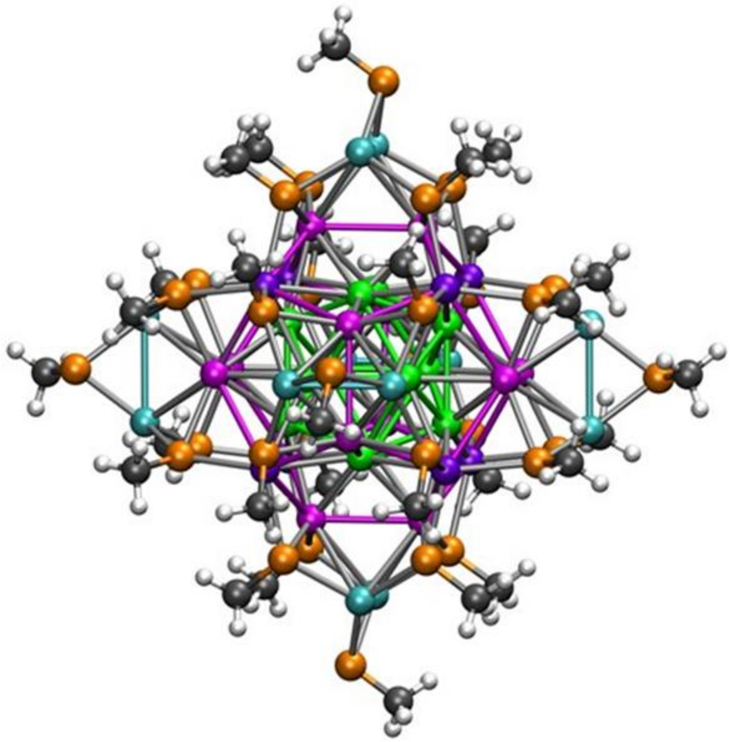

Figure S19. A schematic showing the structure of $Ag_{44}(SR)_{30}$ showing its symmetry unique sites for metal atom substitution reaction. Color codes of atoms: Green (Ag in icosahedral positions), Blue (Ag atoms in the D_{cv} positions), Violet (Ag atoms in the D_{cf} positions), Cyan (Ag atoms in the M positions), Yellow (S atoms of the ligands), Black (C atoms) and White (H atoms).

Supporting Information 20

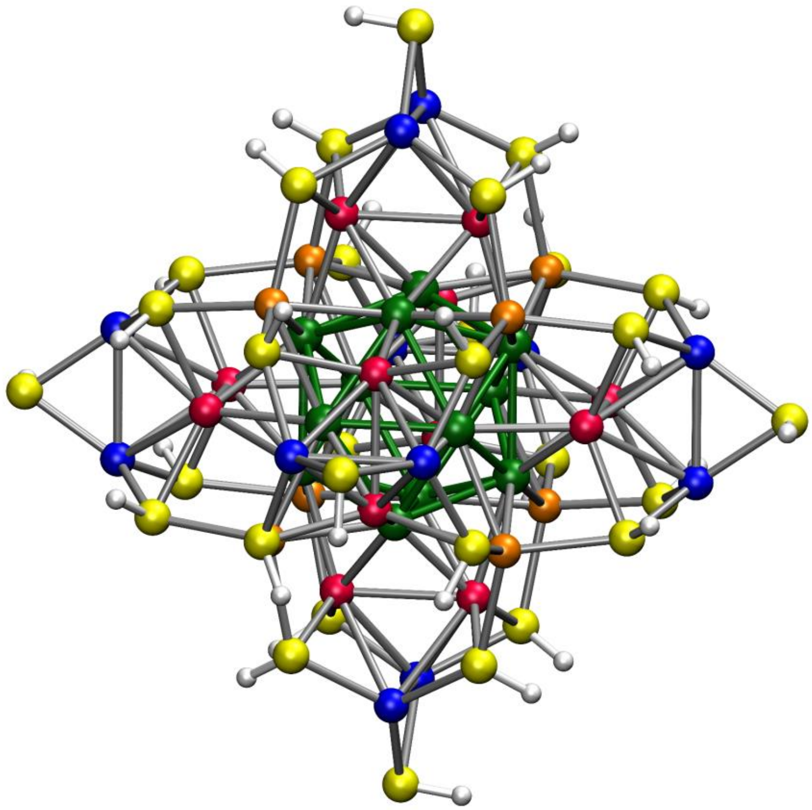

Figure S20. The DFT optimized structure of the D_{cf} isomer of $[Au_{12}Ag_{32}(SR)_{30}]^{4-}$. Color codes for the atoms in the structures: Red (Au), Yellow (S), White (H), Blue (Ag in M positions) and Green (Ag in I position) and Orange (Ag in D_{cv} positions)

Supporting Information 21

Coordinates of the DFT-optimized geometry of the I isomer of $Au_{12}Ag_{32}(SH)_{30}$ in XYZ format.

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Au	10.45240579	12.57137878	9.79803199
Au	13.60632759	11.54205066	14.20616997
Au	12.37113391	10.42494300	9.80610833
Au	11.67508799	13.68115731	14.19949795
Au	13.26859307	13.17024728	9.79954181
Au	10.77625718	10.94050246	14.19519509
Au	10.05730230	10.17948521	11.51415188
Au	13.99159006	13.92687624	12.48872454
Au	14.62960431	11.28583861	11.51590956
Au	9.42163695	12.82460672	12.48125146
Au	11.39462508	14.69563130	11.51444281
Au	12.66807096	9.41759526	12.49601840
Ag	11.35393714	7.81392511	10.47826057
Ag	12.71213937	16.28901588	13.53860903
Ag	16.04054138	13.57807765	10.47996067
Ag	7.99891223	10.53825096	13.51010333
Ag	8.71746108	14.77486142	10.45665245
Ag	15.35926321	9.35203386	13.53845510
Ag	12.03003568	12.04590874	7.44130682
Ag	12.00450314	12.05472236	16.55963750
Ag	9.75559437	10.03659041	8.67989185
Ag	14.28499121	14.08573816	15.32369645
Ag	14.90254759	11.09586478	8.67090150
Ag	9.13820472	13.01258218	15.32567874
Ag	11.41397518	15.04204843	8.68239933
Ag	12.62783251	9.09667160	15.33712359
Ag	7.83616433	11.61114327	10.43276461
Ag	16.21910404	12.51209233	13.55800338
Ag	14.51361222	8.64339683	10.44783363
Ag	9.53420792	15.46431275	13.55048368
Ag	13.74558084	15.92151237	10.44702709
Ag	10.32940368	8.18196436	13.55168737
Ag	7.26176594	11.34887486	7.44583996
Ag	16.75516892	12.75745079	16.54801955
Ag	15.00364355	8.29812182	7.44989353
Ag	9.02746794	15.82673984	16.53068737
Ag	13.80823336	16.51322465	7.46024897
Ag	10.24103956	7.58065309	16.53803898
Ag	6.90481260	9.00523147	9.14947735
Ag	17.13185057	15.11593780	14.85474492
Ag	17.22234936	9.15326247	9.15982239
Ag	6.82064933	14.94119739	14.81875321
Ag	11.96011542	18.02929639	9.14966769
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S	17.12098951	10.63145969	15.16001606
S	13.32052944	6.94386313	8.82580893
S	10.72294448	17.17725639	15.15352051
S	15.82106480	15.72408548	8.84155828
S	8.24424601	8.37566575	15.13044486
S	6.32586201	9.75111757	11.52755650
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S	7.15577004	15.77810335	12.42606930
S	12.82734907	18.12874822	11.55257932
S	11.26809198	5.97013511	12.47870669
S	5.47950610	9.57664755	7.13504837
S	18.53651601	14.52903173	16.88341628
S	17.42820417	7.63956154	7.13124184
S	6.59893236	16.48673658	16.81892812
S	13.17778528	18.94730682	7.12648970
S	10.89810005	5.16292184	16.86327053
H	9.25251030	7.23701185	7.76921937
H	14.75788653	16.86166815	16.26562338
H	17.57152432	12.05510106	7.73563853
H	6.46250800	12.04110730	16.26214248
H	9.25442107	16.88767326	7.75967212
H	14.81570843	7.35628420	16.32560558
H	9.71808725	10.13628401	5.68759826
H	14.30613807	13.95557668	18.32572929
H	14.81325695	11.00449705	5.67803276
H	9.24384714	13.12922539	18.32669814
H	11.54069536	14.99816787	5.68187591
H	12.45956586	9.13001065	18.33664810
H	18.36120660	10.74416185	14.58214375
H	14.01607043	5.90790070	9.39753352
H	10.03825635	18.21818938	14.57630797
H	16.38381505	16.84550257	9.39716074
H	7.69067855	7.25511814	14.56236471
H	5.23309591	10.57665662	11.43167133
H	18.80855864	13.62068661	12.54967421
H	16.74230228	6.92656529	11.45230873
H	7.28796096	17.14069470	12.52900358
H	14.06703787	18.71393837	11.48067067
H	6.20049420	8.82310169	6.24418866
H	17.81029677	15.28754969	17.76656775
H	17.73070767	8.64659396	6.25159274
H	6.28266719	15.49324854	17.71141694

H	12.15279809	18.71246142	6.24675270
H	11.90133295	5.40346821	17.76612653
H	10.04118012	5.36286268	12.58026065
H	5.74296871	13.44920225	9.36953575

Supporting Information 22

Coordinates of the DFT-optimized geometry of the M isomer of $Au_{12}Ag_{32}(SH)_{30}$ in XYZ format.

104

Ag	10.42796687	12.49396514	9.82954371
Ag	13.57162273	11.50267388	14.17147874
Ag	12.33958752	10.38142879	9.82773960
Ag	11.66158230	13.61738344	14.17181137
Ag	13.20943161	13.10736124	9.81550838
Ag	10.79020157	10.89071498	14.18474481
Ag	10.05890760	10.13459577	11.52016791
Ag	13.94063355	13.86489531	12.48007339
Ag	14.55833017	11.24673838	11.51159618
Ag	9.44352461	12.75338272	12.48865801
Ag	11.37836665	14.61195970	11.51042720
Ag	12.62392888	9.38658382	12.48748508
Ag	11.32495808	7.75440208	10.48591174
Ag	12.67441135	16.24694869	13.51019132
Ag	15.97427552	13.53307518	10.46733378
Ag	8.02517600	10.46709197	13.53136886
Ag	8.69608492	14.69765894	10.48326003
Ag	15.31045608	9.30739947	13.50328947
Ag	11.97314746	11.99029393	7.46167932
Ag	12.02172461	12.00970633	16.53870232
Ag	9.75898067	9.96320195	8.69299938
Ag	14.23864711	14.03799638	15.30566456
Ag	14.85869593	11.07923868	8.69398854
Ag	9.14287006	12.92036942	15.30671963
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Ag	12.64924311	9.05948301	15.31514352
Ag	7.83088934	11.56240705	10.47949311
Ag	16.16700808	12.44366438	13.51962258
Ag	14.46039122	8.59807628	10.46207758
Ag	9.55417214	15.40835855	13.53911261
Ag	13.72102448	15.84003697	10.44842592
Ag	10.28950294	8.15281062	13.54988208
Au	7.19782334	11.32772088	7.48347887
Au	16.79942414	12.67727864	16.51580513
Au	14.98739307	8.20407198	7.40570748
Au	9.01054169	15.80216486	16.59311914
Au	13.79912213	16.40940431	7.40549027
Au	10.20319384	7.59387988	16.58965087
Au	6.89516653	8.88450800	9.21171590

Au	17.10355465	15.11829254	14.78942450
Au	17.20958717	9.19123775	9.20070760
Au	6.79619162	14.80392290	14.79634579
Au	11.87527225	17.92960915	9.20249445
Au	12.18742961	6.00707507	14.80142171
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S	6.56315207	12.33713334	14.92204065
S	9.59514464	16.90078828	9.09981592
S	14.41049183	7.10448025	14.90150732
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S	11.39082605	9.55275733	17.61294687
S	6.97075478	13.44679839	8.77412749
S	17.02763269	10.56037216	15.22229249
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S	10.70678026	17.09618678	15.20927861
S	15.77279747	15.62766087	8.74684303
S	8.22963035	8.35584333	15.23412234
S	6.34262769	9.62868935	11.53644158
S	17.65820411	14.37583248	12.46329039
S	16.89433339	8.30053535	11.51355168
S	7.14700035	15.69664093	12.48241533
S	12.79312304	18.10415816	11.50887830
S	11.20791241	5.89954326	12.49581909
S	5.53537064	9.52934125	7.22581559
S	18.46568169	14.46961285	16.77441256
S	17.35217247	7.62273169	7.21382471
S	6.64594077	16.37769811	16.77905911
S	13.10665985	18.78990482	7.19685898
S	10.89537761	5.21004382	16.79548859
H	9.13201980	7.16379036	7.81818611
H	14.85984785	16.84906247	16.17759088
H	17.55212859	11.95232081	7.75844255
H	6.44403143	12.05371042	16.24318512
H	9.26470789	16.87006209	7.78421561
H	14.73880938	7.12948052	16.21803462
H	9.63201864	10.14525044	5.70786765
H	14.36718151	13.85990966	18.28831627
H	14.72291707	10.82962136	5.70228990
H	9.27272983	13.17555046	18.29786161
H	11.56114570	14.96078375	5.68750743
H	12.43684363	9.04555009	18.31242950
H	5.73151171	13.35352638	9.31668410
H	18.26708076	10.65355990	14.68049022
H	13.96929397	5.86837857	9.33809504
H	10.02998824	18.13511901	14.66268465
H	16.30945550	16.74998373	9.28653164
H	7.69087090	7.23230368	14.70073090
H	5.24875260	10.42287185	11.42770803

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H     18.75123765      13.58001539      12.56852389
H     16.76995600       6.95410007      11.40672248
H      7.23481978      17.04657689      12.58304762
H     14.01799813      18.67537453      11.39195830
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H     17.83756326      15.23148927      17.70333932
H     17.69958056       8.53838597       6.27517516
H      6.30172409      15.46490282      17.72117105
H     12.12469715      18.62761454       6.27701529
H     11.87027062       5.37185288      17.72223990
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Supporting Information 23

Coordinates of the DFT-optimized geometry of the D_{cf} isomer of $Au_{12}Ag_{32}(SH)_{30}$ in XYZ format.

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Ag     10.40508339      12.50273125       9.81570923
Ag     13.60250066      11.52174110      14.20797487
Ag     12.39451480      10.39574445       9.80527332
Ag     11.65812652      13.66379085      14.18496649
Ag     13.25562478      13.15454177       9.81407094
Ag     10.78625764      10.88340675      14.20597942
Ag     10.06880442      10.13356819      11.52333196
Ag     14.00345469      13.89191115      12.49874618
Ag     14.63238494      11.25921640      11.51999851
Ag      9.41765343      12.78007115      12.50352625
Ag     11.39427256      14.65152611      11.49232652
Ag     12.67302944       9.38027274      12.49118154
Ag     11.34002445       7.77565686      10.46225537
Ag     12.70811313      16.27167102      13.50553652
Ag     16.04764303      13.54199997      10.45048265
Ag      7.99661578      10.47322982      13.50721352
Ag      8.68528550      14.74120894      10.50269537
Ag     15.35900337       9.31030517      13.54540134
Ag     12.02944536      12.05545522       7.46071205
Ag     12.00300625      12.06285565      16.55463488
Au      9.85175458       9.95459978       8.71185702
Au     14.19254523      14.04696609      15.32062035
Au     14.88445829      11.15543072       8.70996528
Au      9.19699217      12.89473564      15.33016459
Au     11.34694261      14.87461509       8.66813797
Au     12.69781163       9.10281252      15.30292793
Au      7.83274625      11.54403968      10.55601324
Au     16.19554510      12.43077361      13.53017546
Au     14.50334214       8.62678834      10.50525821
Au      9.55725659      15.42063563      13.45791863
Au     13.74855015      15.85845944      10.52678824
Au     10.33618070       8.16753736      13.51624712
Ag      7.29705654      11.27117698       7.46925083
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Ag	13.72324491	16.45678646	7.41962852
Ag	10.30128366	7.52219561	16.55716921
Ag	6.89936590	8.95408281	9.14968758
Ag	17.12616775	15.06238357	14.89739210
Ag	17.23178568	9.12695757	9.14557210
Ag	6.84113191	14.97062255	14.81678565
Ag	11.95801794	17.96066106	9.14001411
Ag	12.14100195	6.04886208	14.89015800
S	8.99397986	7.48450887	9.05466662
S	15.02325244	16.50875045	15.00253193
S	17.45827096	11.66805482	9.03478362
S	6.65123181	12.43165190	14.94839437
S	9.66566654	16.84577330	8.98638971
S	14.44504689	7.14918212	14.97303057
S	9.62033213	11.24378368	6.41372403
S	14.43847213	12.75712801	17.63138440
S	13.88896499	10.31202569	6.41214558
S	10.16662565	13.73371290	17.68262366
S	12.60145469	14.46274651	6.32123889
S	11.49305173	9.54795013	17.58618558
S	6.97244694	13.39090041	8.89855988
S	17.10720981	10.59316954	15.16613543
S	13.36564604	6.92619717	8.87545501
S	10.67711166	17.08415285	15.12973556
S	15.74480366	15.72160562	8.85626376
S	8.29442628	8.34330493	15.15184299
S	6.29167524	9.63572132	11.55303100
S	17.67554462	14.32743956	12.48534378
S	16.89522523	8.28235825	11.55415506
S	7.14760644	15.86523699	12.42929137
S	12.84638880	18.13433737	11.54311703
S	11.22831665	5.93923748	12.47776371
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S	18.66711708	14.39591072	16.80834990
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S	6.63769323	16.58689040	16.76569187
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S	10.79141050	5.04712576	16.79159676
H	9.19465959	7.18290680	7.73031408
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H	6.41918138	12.14249076	16.27075856
H	9.30509217	16.85734730	7.66095241
H	14.81369102	7.20144228	16.29456910
H	9.69546901	10.09481954	5.66410355
H	14.35870071	13.92697210	18.34643600
H	14.84559733	10.96618252	5.67485952
H	9.18837194	13.05084460	18.36237813
H	11.52361884	14.96781364	5.63762187
H	12.52759934	9.04386321	18.33687635
H	18.37590120	10.66255676	14.64577777

H	14.04752116	5.85530175	9.39995017
H	9.98965988	18.16402187	14.63182877
H	16.35621861	16.84981160	9.34574404
H	7.67576678	7.23160481	14.63623266
H	5.17746857	10.43134926	11.43769266
H	18.82253143	13.57611057	12.56518067
H	16.77910140	6.91850759	11.45254066
H	14.08130486	18.72526337	11.43946909
H	10.00063455	5.32952892	12.57202820
H	5.99072680	8.84311892	6.26676720
H	18.06450378	15.17887634	17.75877971
H	17.81246826	8.35339465	6.27969397
H	6.20215396	15.66897565	17.68753841
H	12.30828691	18.85699673	6.26476700
H	11.76274075	5.17422023	17.75100367
H	7.33056335	17.22219964	12.54509441
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