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Unravelling the quantum-entanglement effect of noble gas coordination on the spin ground state of CUO

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Supporting Information

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DMRG occupation numbers

Table I: DMRG occupation numbers for $CUONe_4$ and $CUOAr_4$ molecules in all spin-states.

Orbital index	(1) $^1CUONe_4$	(2) $^3CUO^{(v)}Ne_4$	(3) $^3CUO^{(a)}Ne_4$	(4) $^1CUOAr_4$	(5) $^3CUO^{(v)}Ar_4$	(6) $^3CUO^{(a)}Ar_4$	Δ (1)-(3)	Δ (4)-(6)	Δ (1)-(4)	Δ (3)-(6)
1	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
3	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
4	1.986	1.982	1.980	1.986	1.981	1.980	0.006	0.006	0.000	0.000
5	1.966	1.958	1.954	1.962	1.954	1.954	0.012	0.008	0.004	0.000
6	1.958	0.994	0.989	1.953	0.993	0.989	0.969	0.964	0.005	0.000
7	0.052	0.039	0.046	0.059	0.045	0.047	0.005	0.012	-0.007	-0.001
8	0.018	0.013	0.016	0.019	0.013	0.016	0.002	0.003	-0.001	0.000
9	0.008	0.008	0.009	0.008	0.008	0.009	-0.001	-0.001	0.000	0.000
10	0.006	0.005	0.004	0.007	0.005	0.005	0.001	0.002	-0.001	0.000
11	0.001	0.003	0.003	0.001	0.003	0.003	-0.002	-0.002	0.000	0.000
12	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000
13	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000
14	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
15	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
16	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
17	1.967	1.968	1.966	1.963	1.965	1.966	0.001	-0.003	0.004	0.000
18	1.938	1.943	1.931	1.935	1.940	1.931	0.007	0.004	0.004	0.001
19	0.060	0.498	0.497	0.063	0.498	0.497	-0.438	-0.435	-0.003	0.000
20	0.032	0.053	0.063	0.036	0.055	0.064	-0.031	-0.028	-0.004	0.000
21	0.001	0.033	0.035	0.002	0.037	0.035	-0.034	-0.034	0.000	0.000
22	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000
23	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
24	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
25	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
26	1.967	1.968	1.966	1.963	1.965	1.966	0.001	-0.003	0.004	0.000
27	1.938	1.943	1.931	1.935	1.940	1.931	0.007	0.004	0.004	0.001
28	0.060	0.498	0.497	0.063	0.498	0.497	-0.438	-0.435	-0.003	0.000
29	0.032	0.053	0.063	0.036	0.055	0.064	-0.031	-0.028	-0.004	0.000
30	0.001	0.033	0.035	0.002	0.037	0.035	-0.034	-0.034	0.000	0.000
31	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.000	0.000	0.000
32	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
33	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
34	2.000	2.000	2.000	2.000	2.000	2.000	0.000	0.000	0.000	0.000
35	0.003	0.005	0.005	0.004	0.005	0.005	-0.002	-0.002	-0.001	0.000
36	0.001	0.002	0.002	0.001	0.002	0.002	-0.001	-0.001	0.000	0.000

Electronic energies for the dissociation of Ng_4 (Ng = Ne, Ar) from CUO

Table II: Energies for the $^1\Sigma^+$ and $^3\Phi$ states of $CUONe_4$ and $CUOAr_4$ in Hartree atomic units for DMRG(38,36)[m_{start},m_{min} = 512, m_{max},10^{-5}] calculations for different interatomic distances d_{UNg} (Ng = Ne, Ar).

$CUONe_4$	E/Hartree (−28621.)										
$^1\Sigma^+$	2.83 Å	3.11 Å	3.30 Å	3.54 Å	3.89 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	8.46 Å	14.14 Å
CAS(12,12)SCF	780 576	788 844	791 039	792 098	792 692	792 817	792 806	792 763	792 682	792 606	792 606
DMRG(38,36)											
[1024,512,1024,10^{-5}]	798 573	806 894	809 271	810 565	811 330	811 518	811 424	811 337	811 121	810 948	810 875
[2048,512,2048,10^{-5}]	798 591	806 906	809 283	810 577	811 343	811 527	811 441	811 346	811 130	810 968[a]	810 909[a]

$CUONe_4$	E/Hartree (−28621.)										
$^3\Phi$ vertical	2.83 Å	3.11 Å	3.30 Å	3.89 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	6.36 Å	7.07 Å	14.14 Å
CAS(12,14)SCF	739 079	746 721	748 439	749 224	749 121	749 005	748 917	748 811	748 761	748 741	748 732
DMRG(38,36)											
[1024,512,1024,10^{-5}]	764 563	771 638	773 161	773 636	773 404	773 177	773 009	772 810	772 710	772 675	772 267
[2048,512,2048,10^{-5}]	764 588	771 656	773 178	773 639	773 407	773 184	773 014	772 813	772 723	772 683	772 667

$CUONe_4$	E/Hartree (−28621.)										
$^3\Phi$ adiabatic	2.83 Å	3.11 Å	3.30 Å	3.54 Å	3.89 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	8.46 Å	14.04 Å
CAS(12,14)SCF	759 413	766 461	767 862	768 439	768 481	768 339	768 209	768 115	768 006	767 928	767 928
DMRG(38,36)											
[1024,512,1024,10^{-5}]	786 175	792 656	793 861	794 270	794 129	793 832	793 578	793 391	793 172	793 021	793 017
[2048,512,2048,10^{-5}]	786 202	792 677	793 892	794 289	794 155[a]	793 876[a]	793 590	793 404	793 215	793 052	793 037

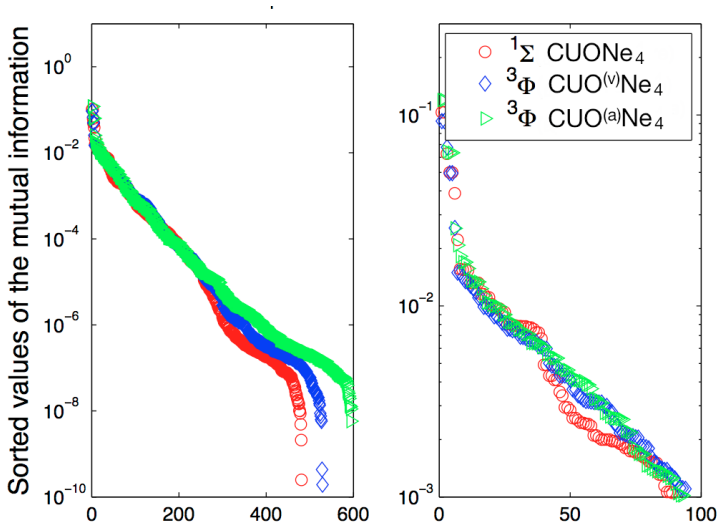
$CUOAr_4$	E/Hartree (−30221.)										
$^1\Sigma^+$	2.83 Å	3.11 Å	3.41 Å	4.06 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	6.36 Å	7.07 Å	14.14 Å
CAS(12,12)SCF	528 701	563 968	575 134	578 519	578 502	578 335	578 163	577 944	577 834	577 783	577 750
DMRG(38,36)											
[1024,512,1024,10^{-5}]	549 848	583 936	594 946	598 645	598 647	598 404	598 050	597 550	597 247	597 169	597 115
[2048,512,2048,10^{-5}]	549 920	584 015	594 974	598 671	598 690	598 428	598 079	597 565	597 284	597 181	597 126

$CUOAr_4$	E/Hartree (−30221.)										
$^3\Phi$ vertical	2.83 Å	3.11 Å	3.41 Å	3.77 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	6.36 Å	7.07 Å	14.14 Å
CAS(12,14)SCF	492 900	528 301	539 203	541 725	541 636	541 300	541 052	540 791	540 681	540 632	540 604
DMRG(38,36)											
[1024,512,1024,10^{-5}]	521 848	555 598	565 566	567 515	566 950	566 330	565 858	565 324	565 090	565 003	564 959
[2048,512,2048,10^{-5}]	521 907	555 642	565 604	567 560	566 982	566 350	565 869	565 330	565 105	565 014	564 967

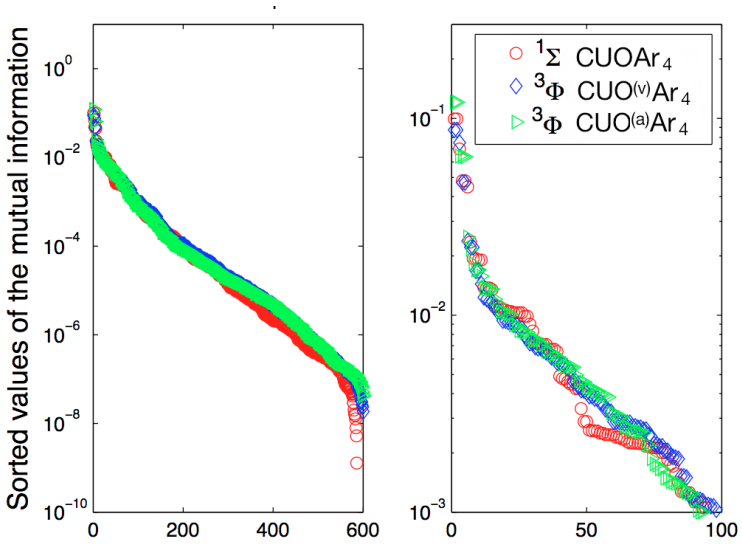
$CUOAr_4$	E/Hartree (−30221.)										
$^3\Phi$ adiabatic	2.83 Å	3.11 Å	3.31 Å	3.75 Å	4.24 Å	4.60 Å	4.95 Å	5.66 Å	6.36 Å	7.07 Å	14.14 Å
CAS(12,14)SCF	508 046	543 398	551 961	556 578	556 474	556 126	555 872	555 606	555 495	555 447	555 420
DMRG(38,36)											
[1024,512,1024,10^{-5}]	537 992	571 597	579 429	583 167	582 526	581 892	581 424	580 903	580 686	580 602	580 558
[2048,512,2048,10^{-5}]	538 048	571 639	579 467	583 200	582 549	581 910	581 439	580 911	580 698	580 614	580 582

[a] m_{min} had to be increased to 768 renormalize active-system states due to convergence difficulties.

Additional entanglement diagrams.

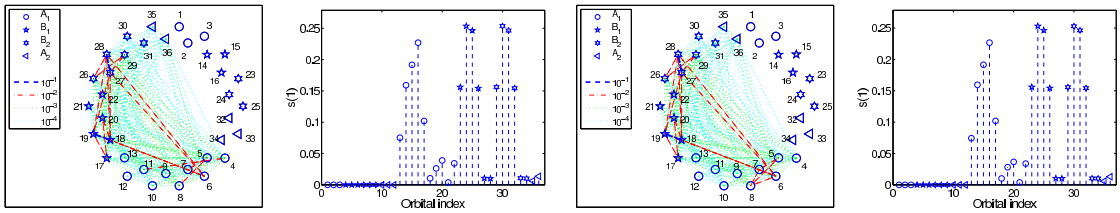


(a) CUONe$_4$



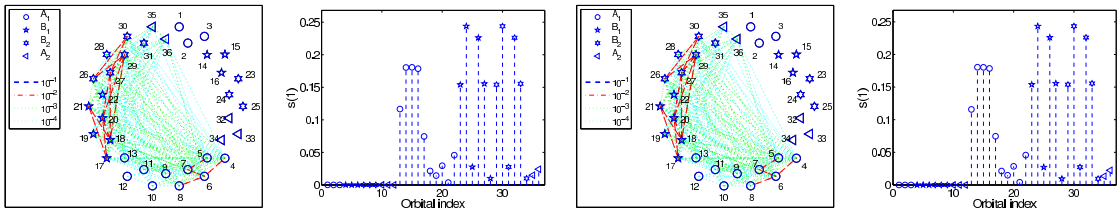
(b) CUOAr$_4$

Figure 1: Decay of mutual information in the CUONg$_4$ molecules in different spin states (around their equlibrium geometries).



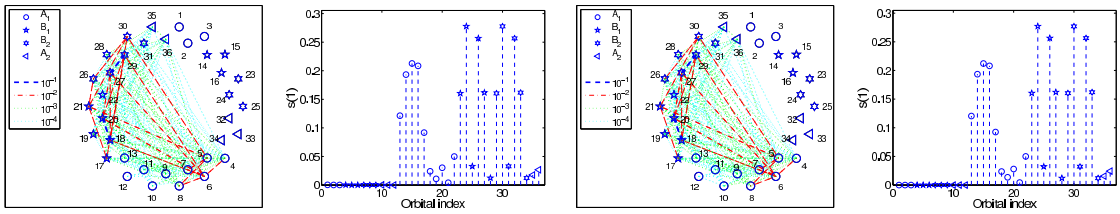
(i) 4.24 Å (ii) 14.14 Å

(a) $CUONe_4$, $^1\Sigma^+$



(i) 3.89 Å (ii) 14.14 Å

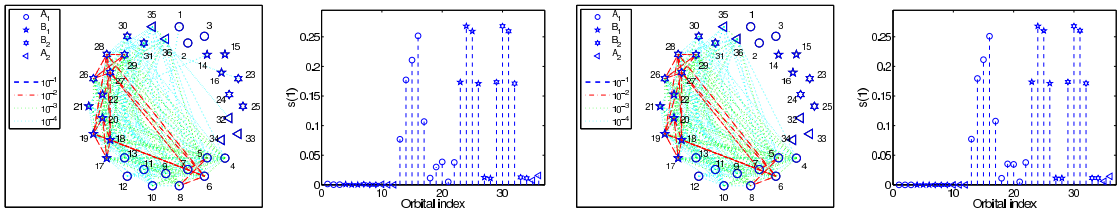
(b) $CUONe_4$, $^3\Phi_{(v)}$



(i) 3.54 Å (ii) 14.14 Å

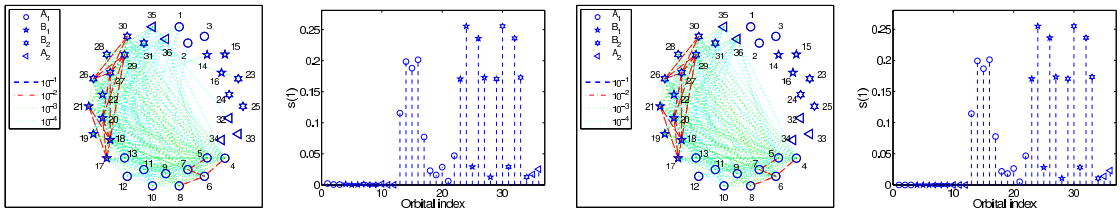
(c) $CUONe_4$, $^3\Phi_{(a)}$

Figure 2: Mutual information and single orbital entropy $s(1)$ for $CUONe_4$ determined at the equilibrium distance and in the vicinity of dissociation of the noble gas matrix. $^3\Phi_{(v)}$: $^3\Phi$ state for $CUO^{(v)}Ng_4$. $^3\Phi_{(a)}$: $^3\Phi$ state for $CUO^{(a)}Ng_4$.



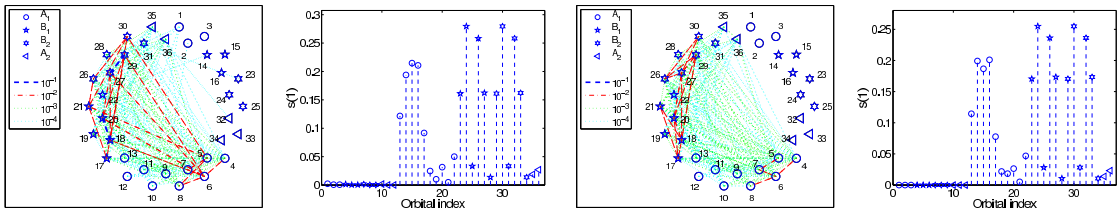
(i) 4.06 Å (ii) 14.14 Å

(a) $CUOAr_4$, $^1\Sigma^+$



(i) 3.77 Å (ii) 14.14 Å

(b) $CUOAr_4$, $^3\Phi_{(v)}$



(i) 3.75 Å (ii) 14.14 Å

(c) $CUOAr_4$, $^3\Phi_{(a)}$

Figure 3: Mutual information and single orbital entropy $s(1)$ for $CUOAr_4$ determined at the equilibrium distance and in the vicinity of dissociation of the noble gas matrix. $^3\Phi_{(v)}$: $^3\Phi$ state for $CUO^{(v)}Ng_4$. $^3\Phi_{(a)}$: $^3\Phi$ state for $CUO^{(a)}Ng_4$.