Supporting Information
for
Bonding of C_1 fragments on metal nanoclusters: a search for methane conversion catalysts with swarm intelligence

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S1. Energy diagram of dehydrogenation of CH_4 to C in the gas phase

As shown in Figure S1, methane is a very stable compound, and breaking the C-H bonds in the gas phase requires about 5 eV of energy per one. It takes about 20 eV of energy to break all the C-H bonds in CH_4 to generate C. The energy diagram shown in Figure S1 was calculated in the following method. Each of H, C, CH, CH_2, CH_3, and CH_4 was placed in a unit cell of about 20 × 20 × 20 $Å^3$ and the structure was optimized. The calculation conditions were set so that the cutoff energy is 500 eV and the k-point is sampled only at the Γ point. For the pseudopotential, we used the projector augmented wave (PAW) method.[1,2] The convergence condition for self-consistent field (SCF) was set to 1.0×10^{-5} eV and the convergence condition for structural optimization was set to 1.0×10^{-4} eV. Grimme's DFT-D2 method was used as a dispersion force correction.[3] The calculations were performed taking spin polarization into account.

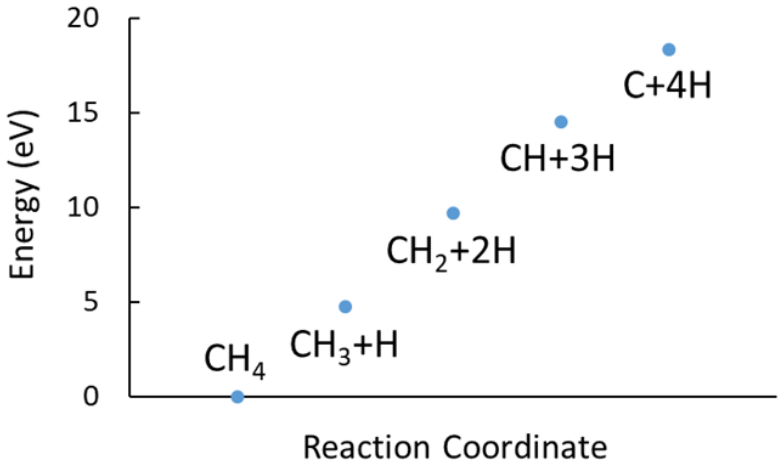


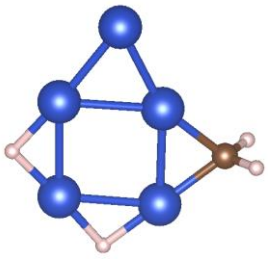
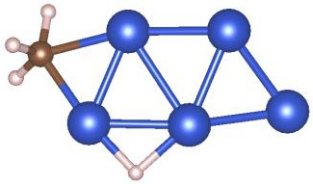
Figure S1. Energy diagram for the sequential dehydrogenation of CH_4 to C in the gas phase.

S2. Effects of dipole correction

For systems with a net dipole moment, due to the periodic boundary condition, not only the total energy converges slowly with respect to the size of the unitcell, but also the potential and the forces are affected by finite size errors. It would be better to check how much of an impact it has, so we compare calculations with and without dipole correction using the structures of two isomers for Cu_5CH_4.

In a single-point energy calculation for the optimized structures for Cu_5CH_4, corrections to the total energy were included by considering the full dipole moment in all directions. Corrections to the potential and forces were also applied.[4] A comparison of the energies calculated for Cu_5CH_4 with and without dipole correction is shown in Table S1. A close inspection of this table shows that the effect of dipole correction can be deemed not to be so significant. This may be due to the weak polarization of the system.

Table S1. Comparison of the energies calculated for the two isomers of Cu_5CH_4. The geometry of Cu_5CH_4 was optimized without dipole correction. Using the optimized geometry, a single-point energy calculation with dipole correction was performed.

	CH_2-Cu_5	CH_3-Cu_5
Optimized geometry		
Total energy calculated without dipole correction (eV)	-34.4145	-34.4088
Total energy calculated with dipole correction (eV)	-34.4142	-34.4080

S3. Boltzmann distribution at 300K

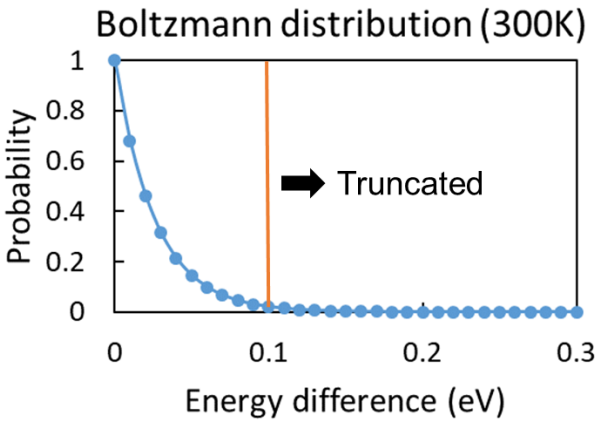


Figure S2. Boltzmann distribution at 300 K. One can see that if the energy difference is larger than 0.1 eV, the probability is negligibly small.

S4. All the cluster structures optimized in this study

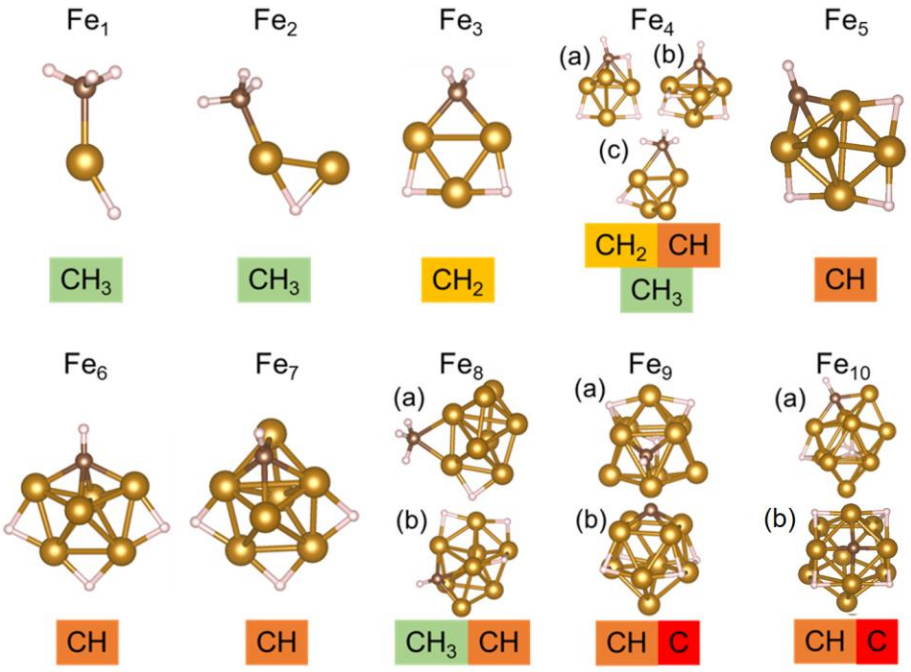


Figure S3. The most stable structure for each of Fe_nCH_4 (n = 1-10) nanoclusters. If a metastable structure exists, the structures are labelled (a), (b), and (c), where (a) indicates the most stable structure while (b) and (c) indicate metastable structures in alphabetical order from the most stable to the least stable. Fe-Fe bonds are shown only when the Fe-Fe bond length is 3.0 Å or less, Fe-C bonds are shown only when the Fe-C bond length is 2.5 Å or less, and Fe-H bonds are shown only when the Fe-H bond length is 2.0 Å or less.

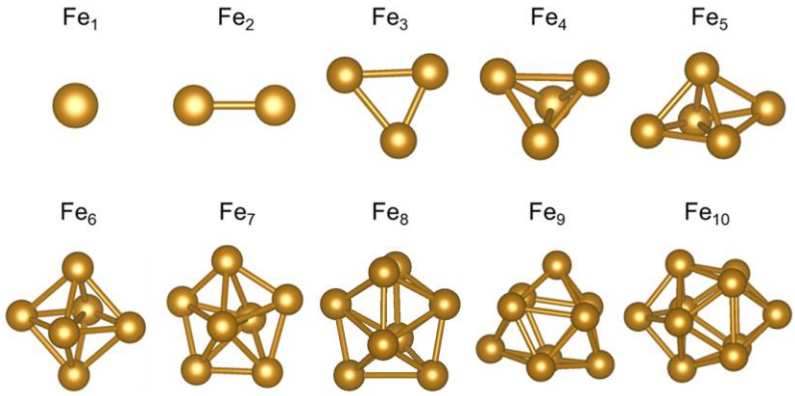


Figure S4. The most stable structure for each of Fe_n (n = 1-10) nanoclusters.

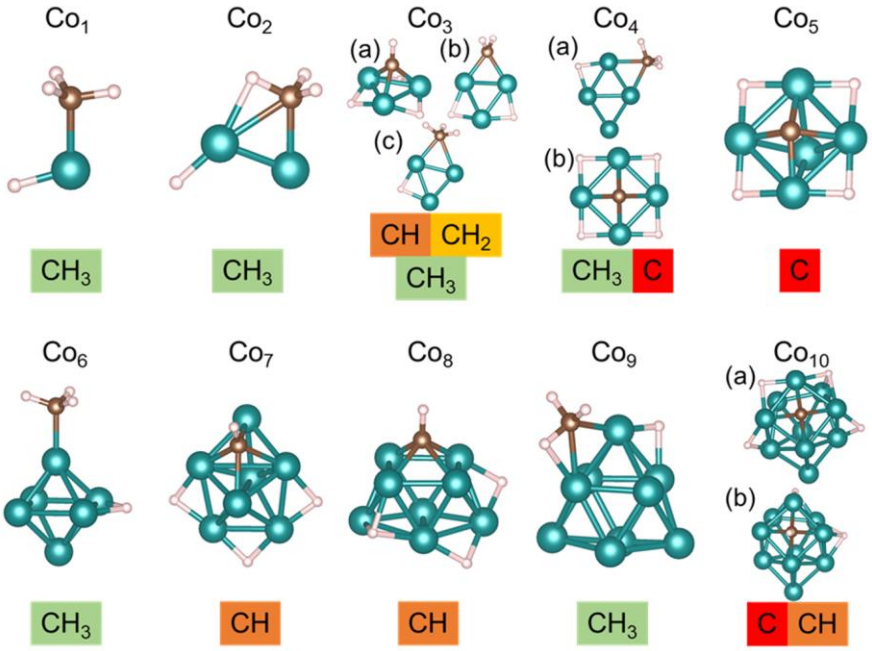


Figure S5. The most stable structure for each of Co_nCH_4 (n = 1-10) nanoclusters. If a metastable structure exists, the structures are labelled (a), (b), and (c), where (a) indicates the most stable structure while (b) and (c) indicate metastable structures in alphabetical order from the most stable to the least stable. Co-Co bonds are shown only when the Co-Co bond length is 3.0 Å or less, Co-C bonds are shown only when the Co-C bond length is 2.5 Å or less, and Co-H bonds are shown only when the Co-H bond length is 2.0 Å or less.

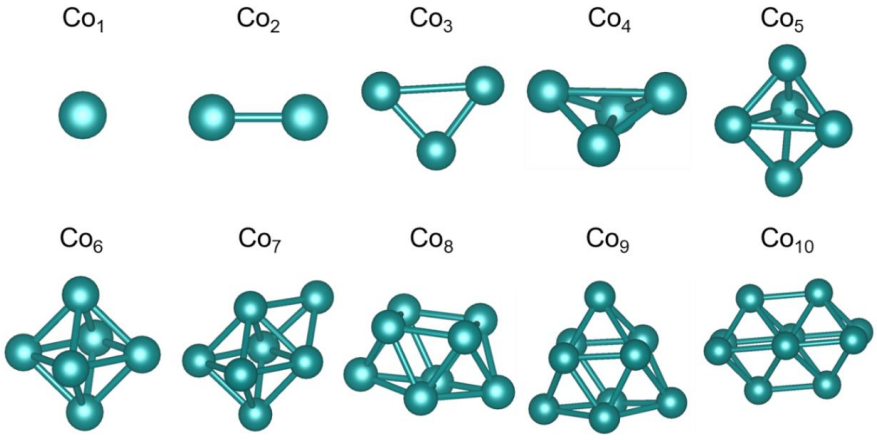


Figure S6. The most stable structure for each of Co_n (n = 1-10) nanoclusters.

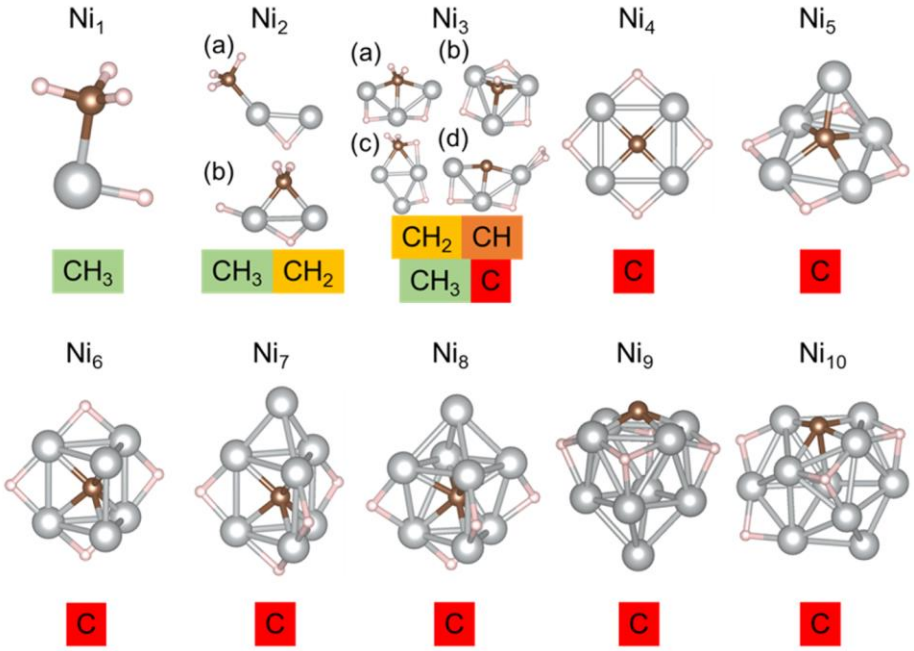


Figure S7. The most stable structure for each of Ni_nCH_4 (n = 1-10) nanoclusters. If a metastable structure exists, the structures are labelled (a), (b), (c) and (d), where (a) indicates the most stable structure while (b), (c), and (d) indicate metastable structures in alphabetical order from the most stable to the least stable. Ni-Ni bonds are shown only when the Ni-Ni bond length is 3.0 Å or less, Ni-C bonds are shown only when the Ni-C bond length is 2.5 Å or less, and Ni-H bonds are shown only when the Ni-H bond length is 2.0 Å or less. Note that the Ni nanoclusters incorporate the C atom inside the clusters when $6 \leq n \leq 8$.

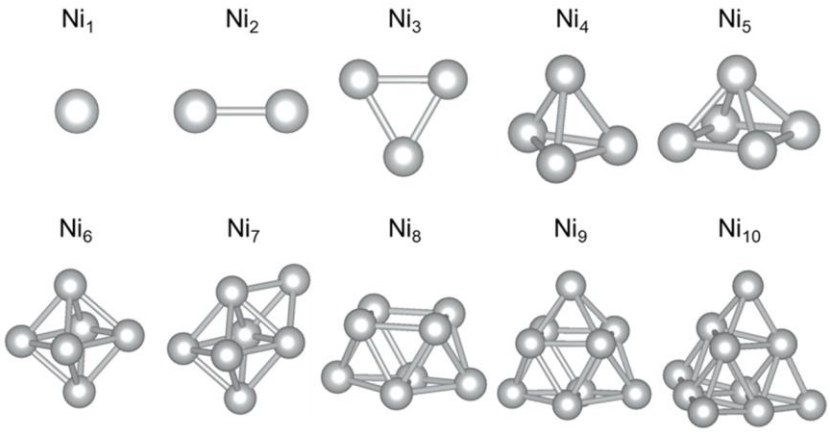


Figure S8. The most stable structure for each of Ni_n (n = 1-10) nanoclusters.

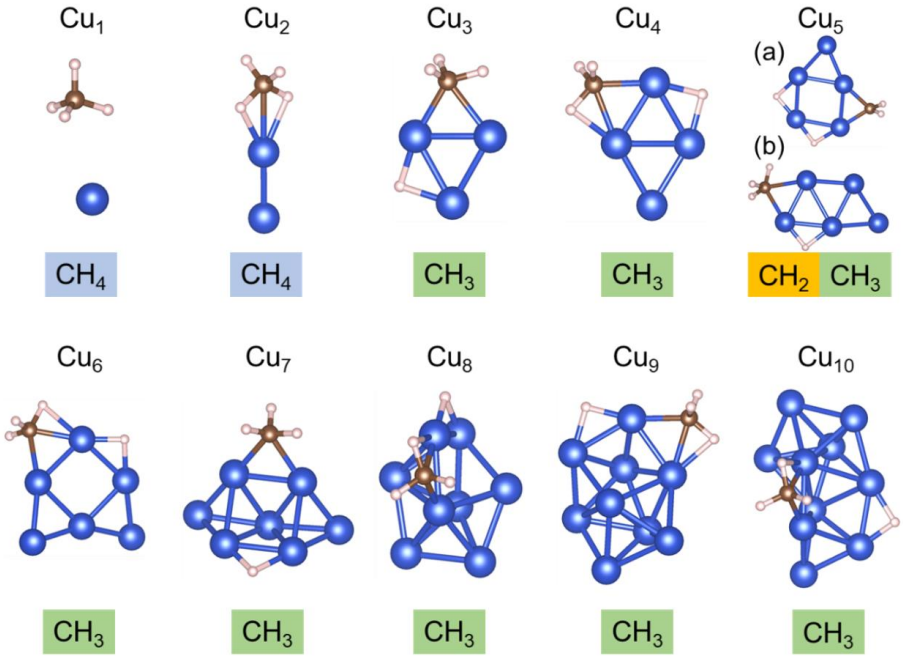


Figure S9. The most stable structure for each of Cu_nCH_4 (n = 1-10) nanoclusters. If a metastable structure exists, the structures are labelled (a) and (b), where (a) indicates the most stable structure while (b) indicates the metastable structure. Cu-Cu bonds are shown only when the Cu-Cu bond length is 3.0 Å or less, Cu-C bonds are shown only when the Cu-C bond length is 2.5 Å or less, Cu-H bonds are shown only when the Cu-H bond length is 2.0 Å or less.

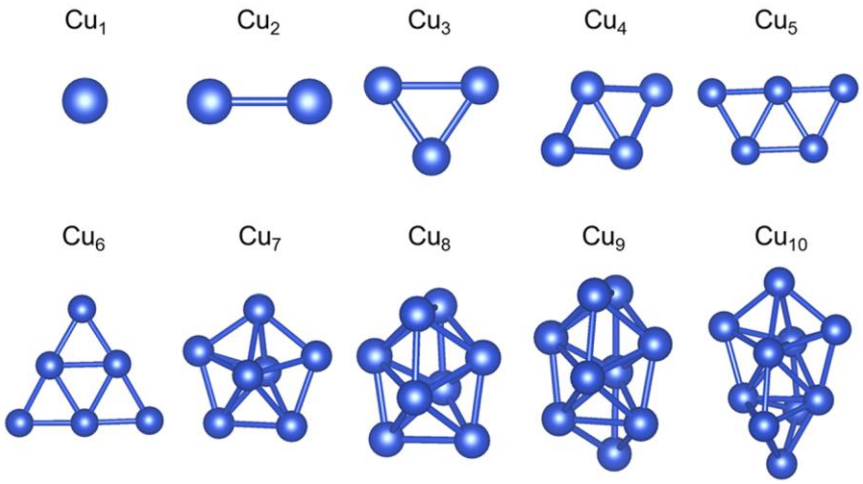


Figure S10. The most stable structure for each of Cu_n (n = 1-10) nanoclusters.

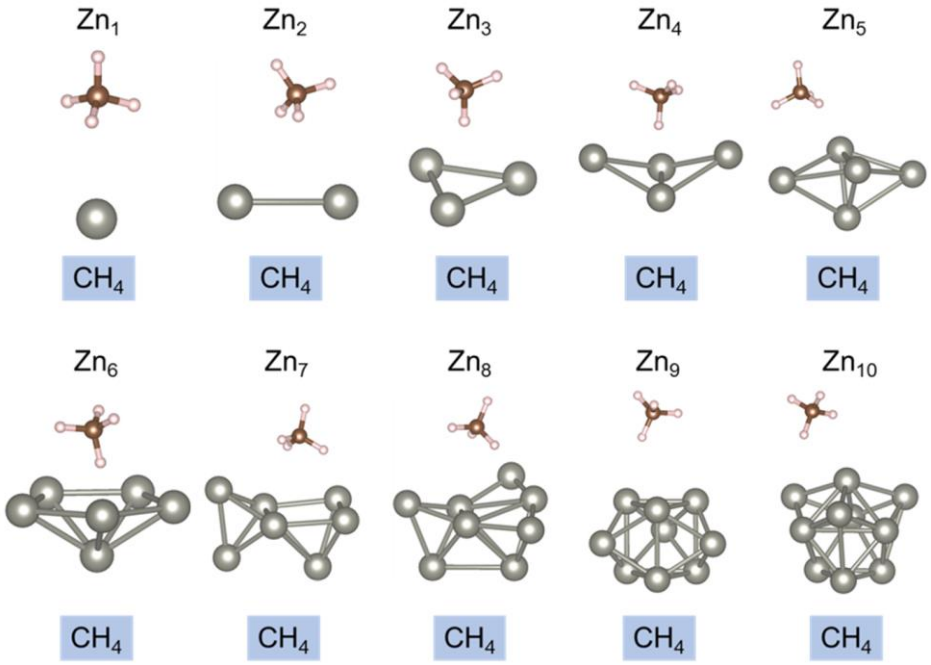


Figure S11. The most stable structure for each of Zn_nCH_4 (n = 1-10) nanoclusters. Zn-Zn bonds are shown only when the Zn-Zn bond is 3.5 Å or less. Note that the Zn nanoclusters cannot activate CH_4.

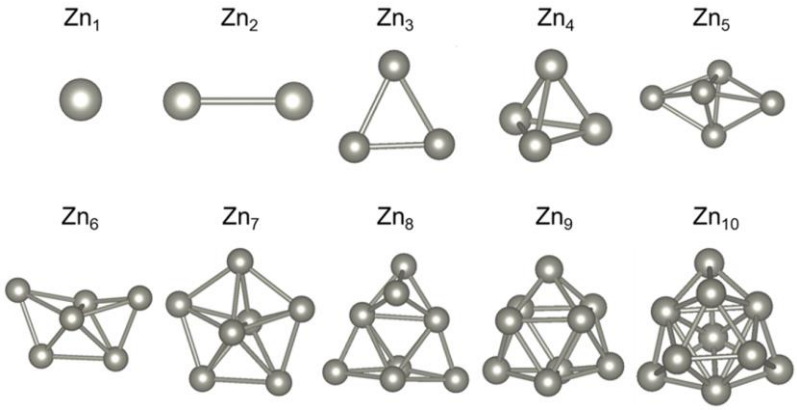


Figure S12. The most stable structure for each of Zn_n (n = 1-10) nanoclusters.

S5. Magnetic moments of all the cluster structures optimized in this study

Table S2. Magnetic moments (in units of μ_B) of the optimized structures shown in Figures S4, S6, S8, S10, and S12.

	M = Fe	M = Co	M = Ni	M= Cu	M = Zn
M_1	4.0	3.0	2.0	1.0	0.0
M_2	6.0	4.2	2.0	0.0	0.0
M_3	10.0	6.8	2.0	1.0	0.0
M_4	14.0	10.0	4.0	0.0	0.0
M_5	16.0	13.0	6.0	1.0	0.0
M_6	20.0	14.0	8.0	0.0	0.0
M_7	22.0	15.0	8.0	1.0	0.0
M_8	18.0 (24.0[a])	16.0	8.0	0.0	0.0
M_9	26.0	17.0	8.0	1.0	0.0
M_{10}	28.0	20.0	8.0	0.0	0.0

[a] This number was obtained when an "all-band simultaneous update of orbitals" conjugate gradient method[4] was adopted as the electronic minimisation algorithm.

Table S3. Magnetic moments (in units of μ_B) of the optimized structures shown in Figures S3, S5, S7, S9, and S11.

	M = Fe	M = Co	M = Ni	M= Cu	M = Zn
M_1CH_4	4.0	1.0	0.0	1.0	0.0
M_2CH_4	6.0	4.0	(a) 2.0 (b) 0.0	0.0	0.0
M_3CH_4	10.0	(a) 3.0 (b) 7.0 (c) 6.7	(a) 0.0 (b) 0.0 (c) 2.0 (d) 0.0	1.0	0.0
M_4CH_4	(a) 12.0 (b) 12.0 (c) 12.0	(a) 8.0 (b) 6.0	0.0	0.0	0.0
M_5CH_4	8.0 (14.0[a])	7.0	2.0	(a) 1.0 (b) 1.0	0.0
M_6CH_4	18.0	12.0	2.0	0.0	0.0
M_7CH_4	20.0	13.0	2.0	1.0	0.0
M_8CH_4	(a) 24.0 (b) 24.0	16.0	2.0	0.0	0.0
M_9CH_4	(a) 26.0 (b) 26.0	17.0	4.0	0.9	0.0
$M_{10}CH_4$	(a) 30.0 (b) 24.1 (28.1[a])	(a) 16.0 (b) 20.0	4.0	0.0	0.0

[a] This number was obtained when an "all-band simultaneous update of orbitals" conjugate gradient method[4] was adopted as the electronic minimisation algorithm.

Using an option to set the difference between the number of electrons in the up and down spin components,[4] we have optimized all of the clusters investigated in this study with the spin multiplicity varied. Then, we have generated a plot of the total energy of the cluster versus the magnetic moment as shown in Figure S13. A comparison of the most stable spin state shown in Figure S13 with the magnetic moments in Tables S2 and S3 shows that except for the clusters of Fe_8, Fe_5CH_4, and $Fe_{10}CH_4$(b), the most stable spin state can be properly predicted by the spin polarization calculations with VASP. We recalculated the Fe_8, Fe_5CH_4, and $Fe_{10}CH_4$(b) clusters with the correct spin multiplicity.

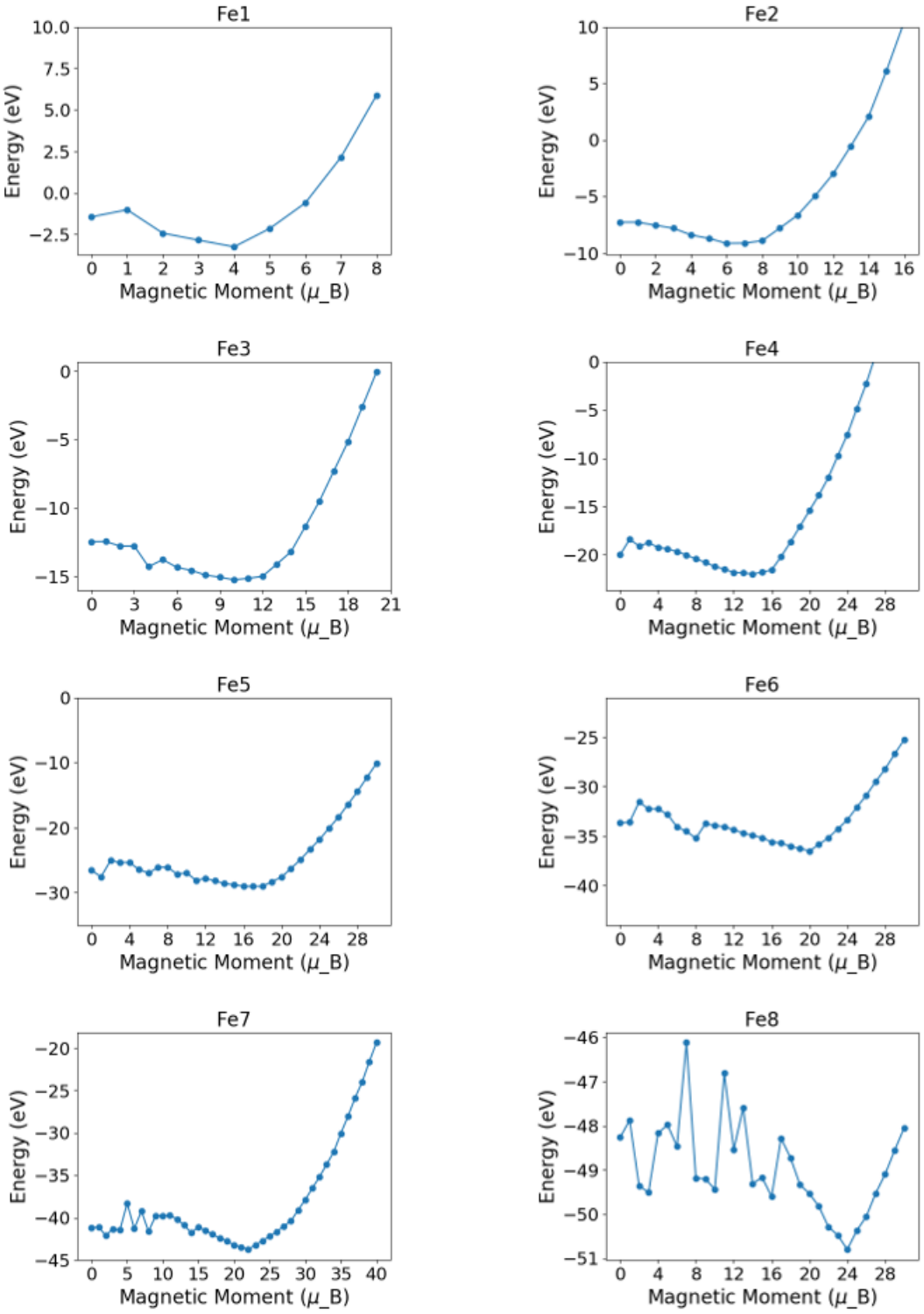


Figure S13. Total energy of each cluster is plotted as a function of the magnetic moment.

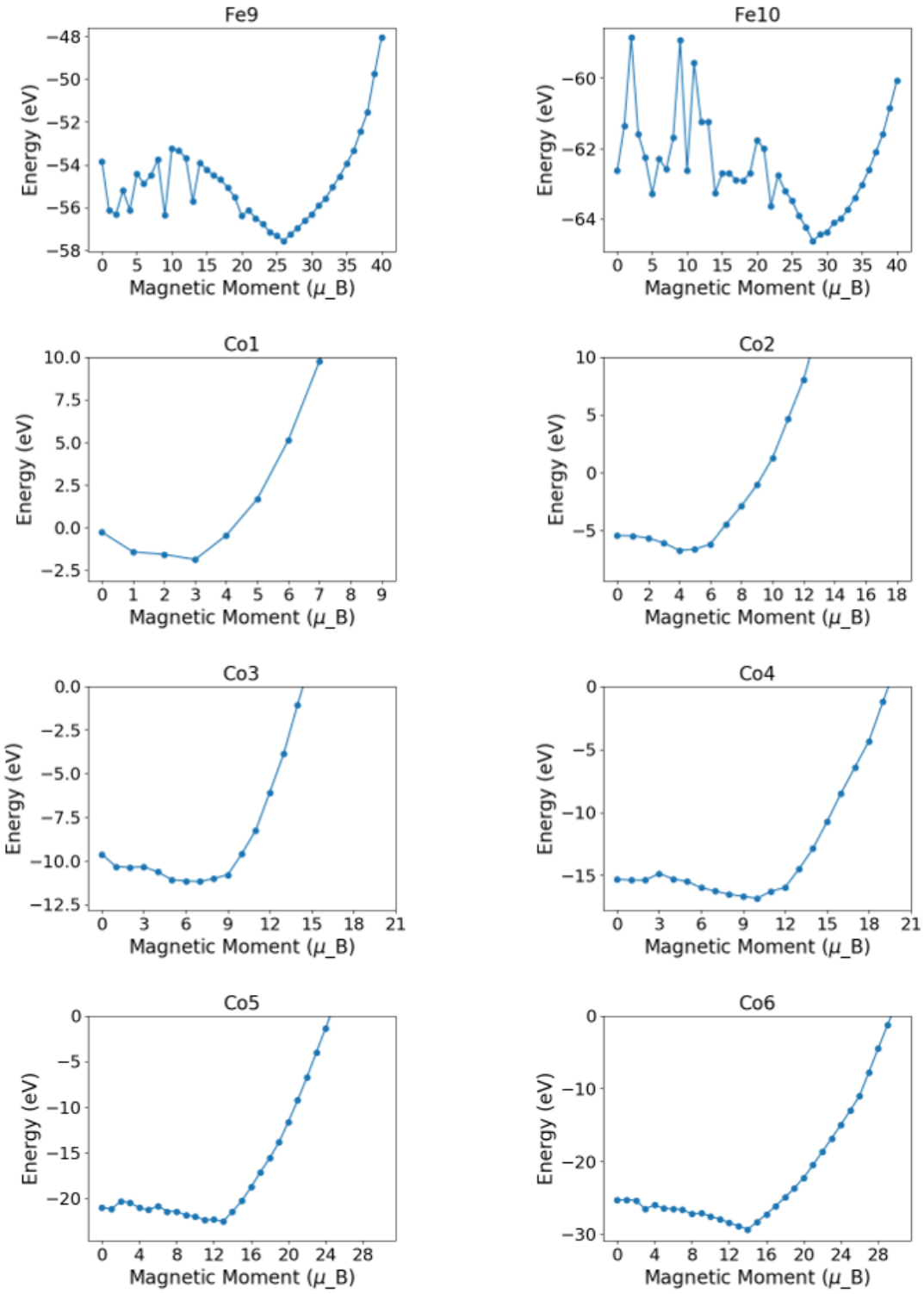
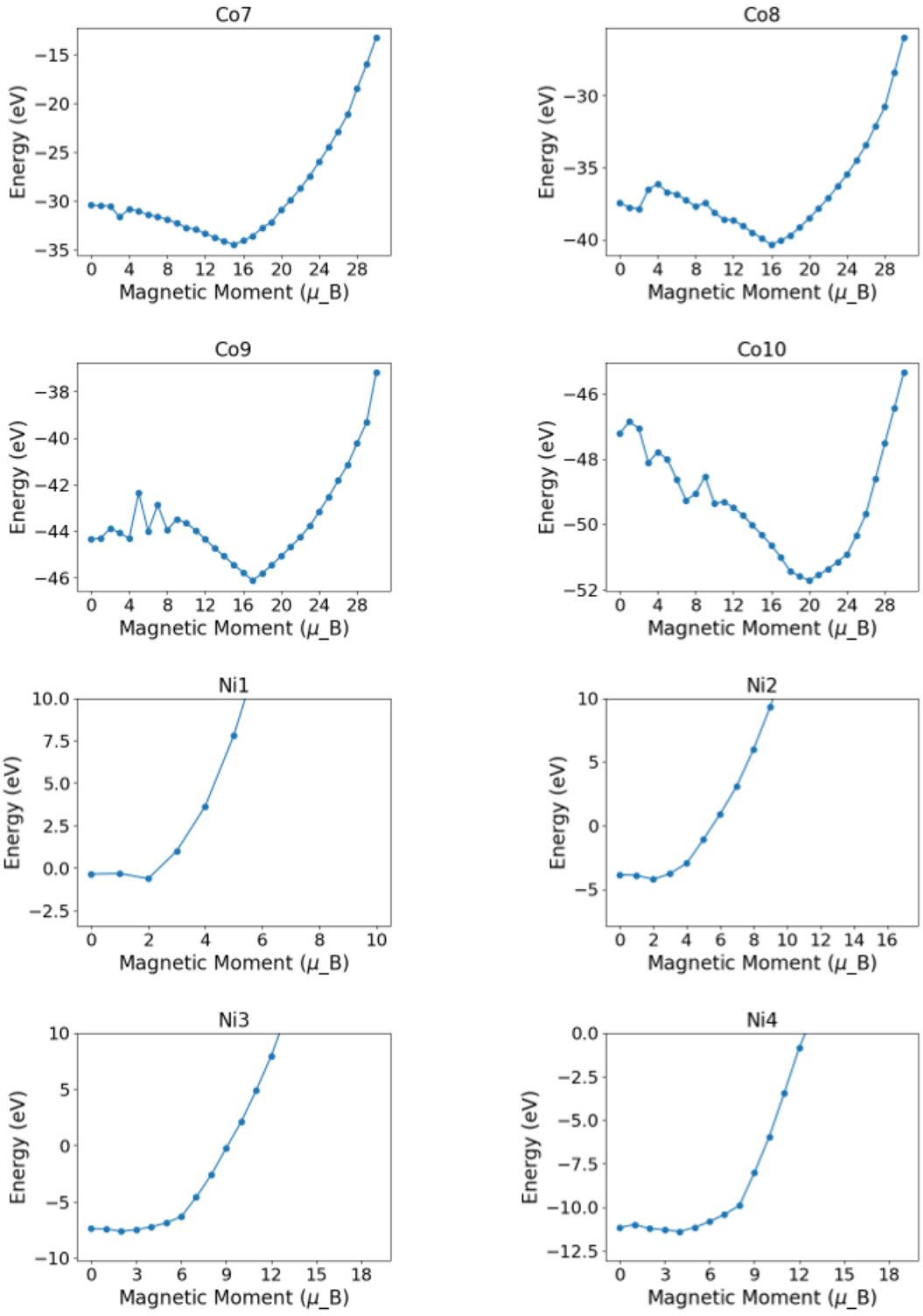


Figure S13. Continued.



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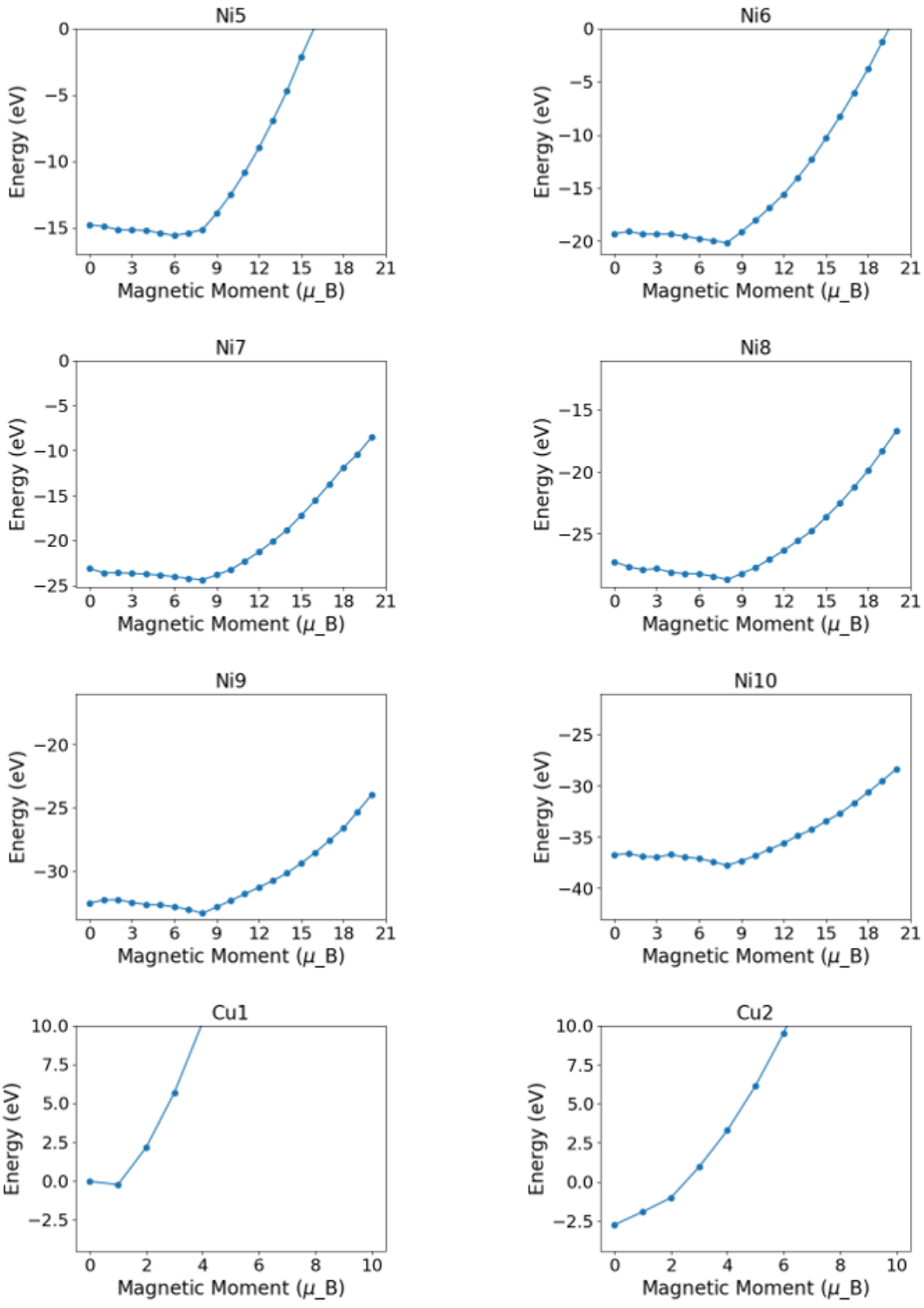

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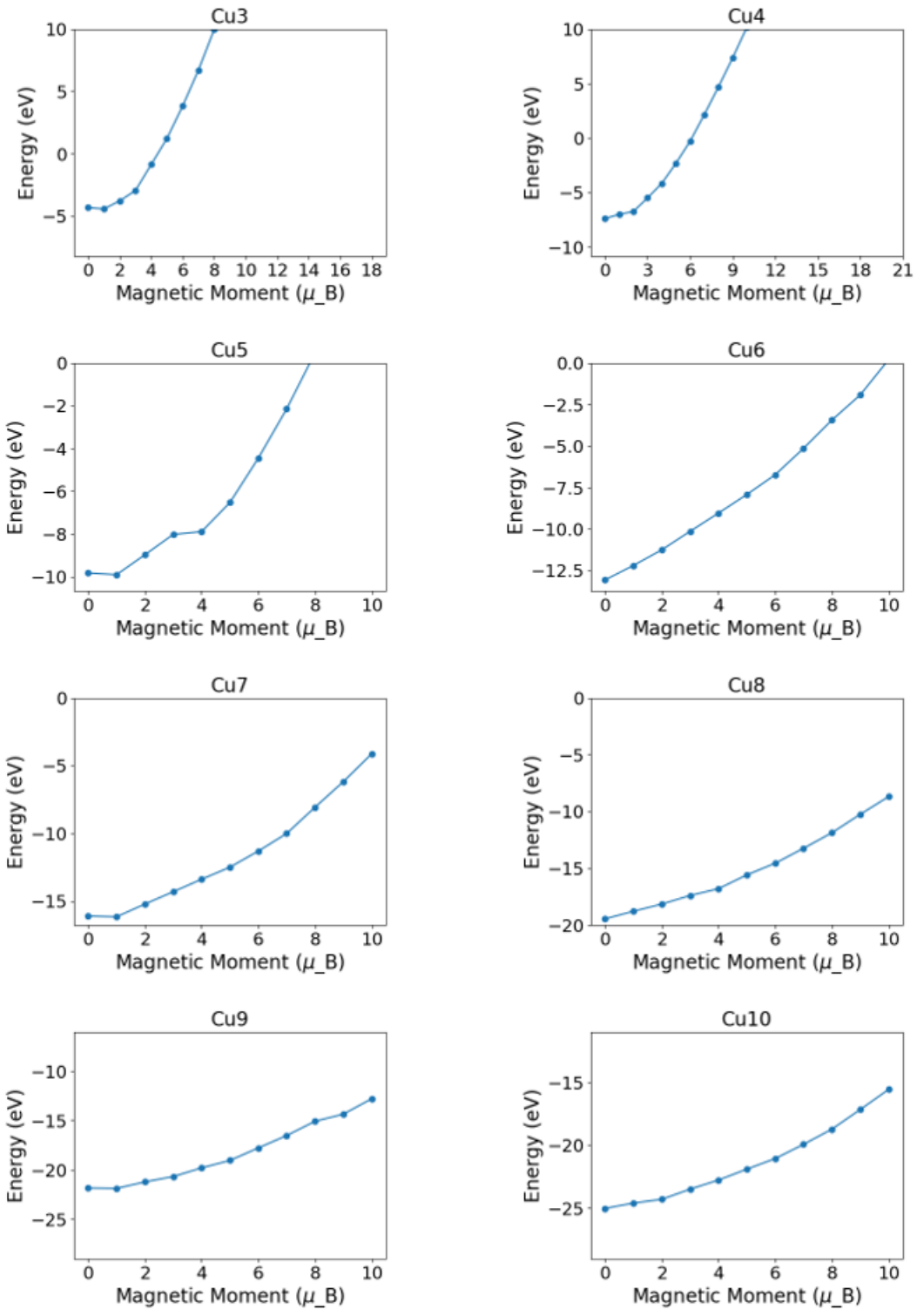

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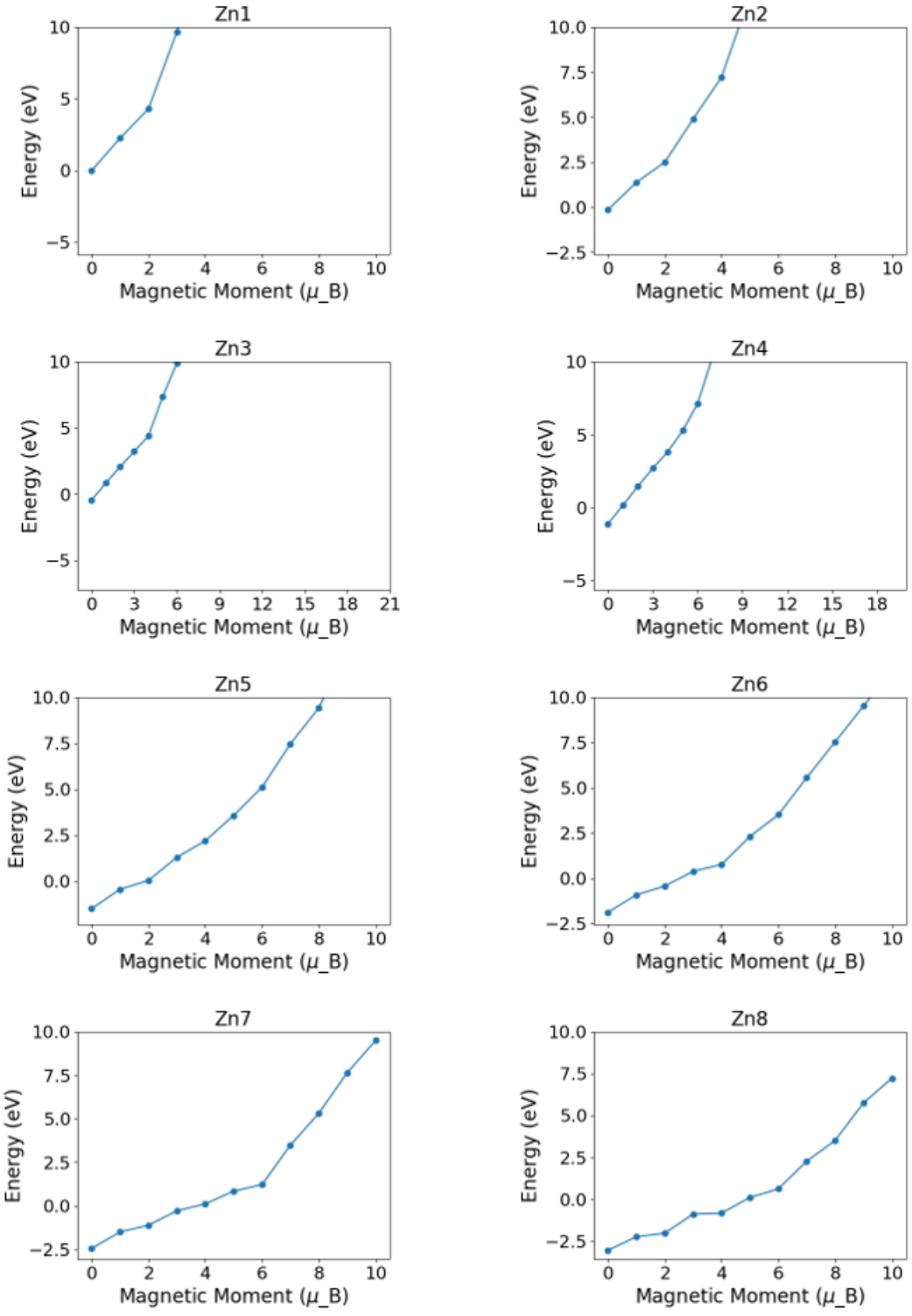

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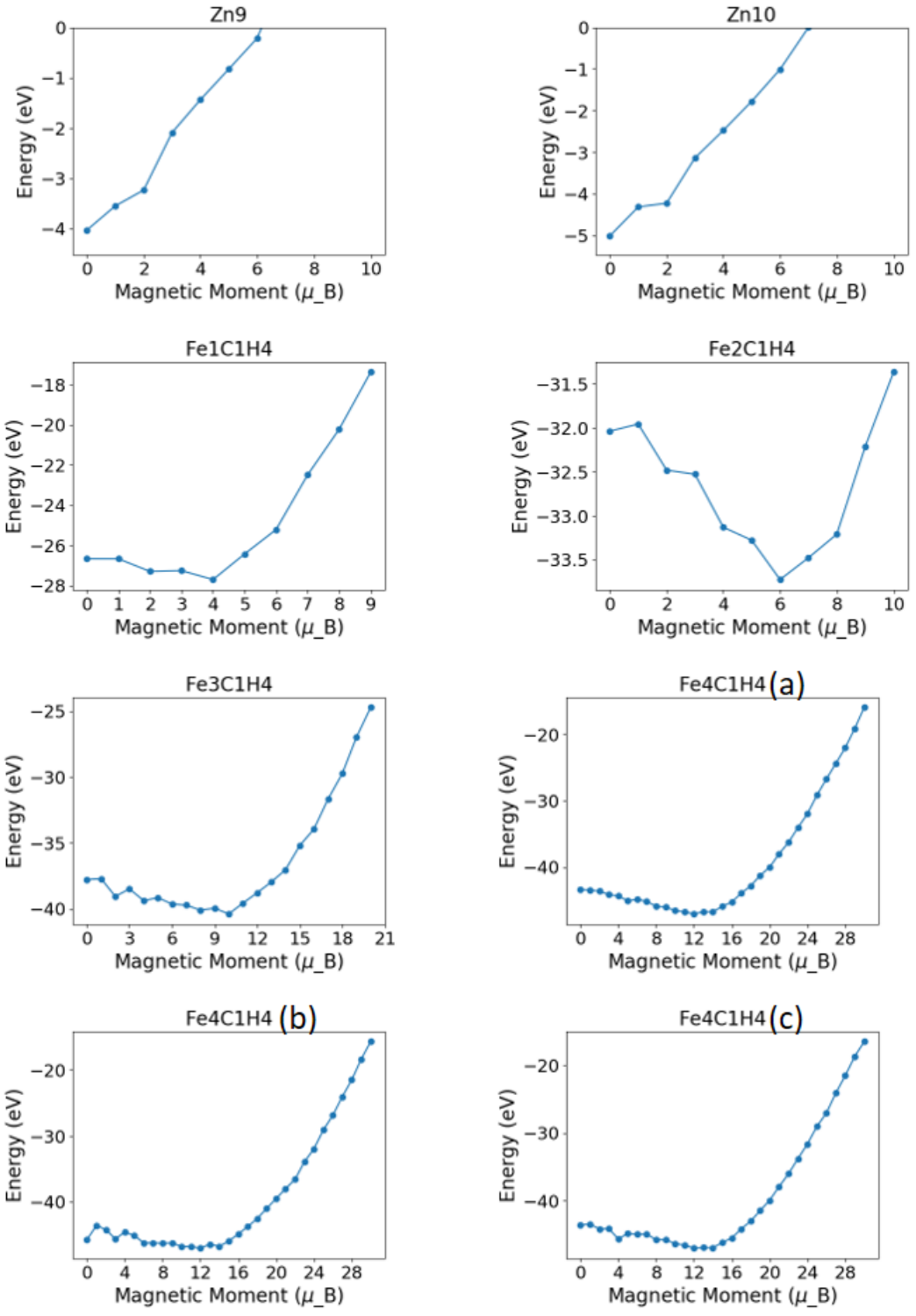

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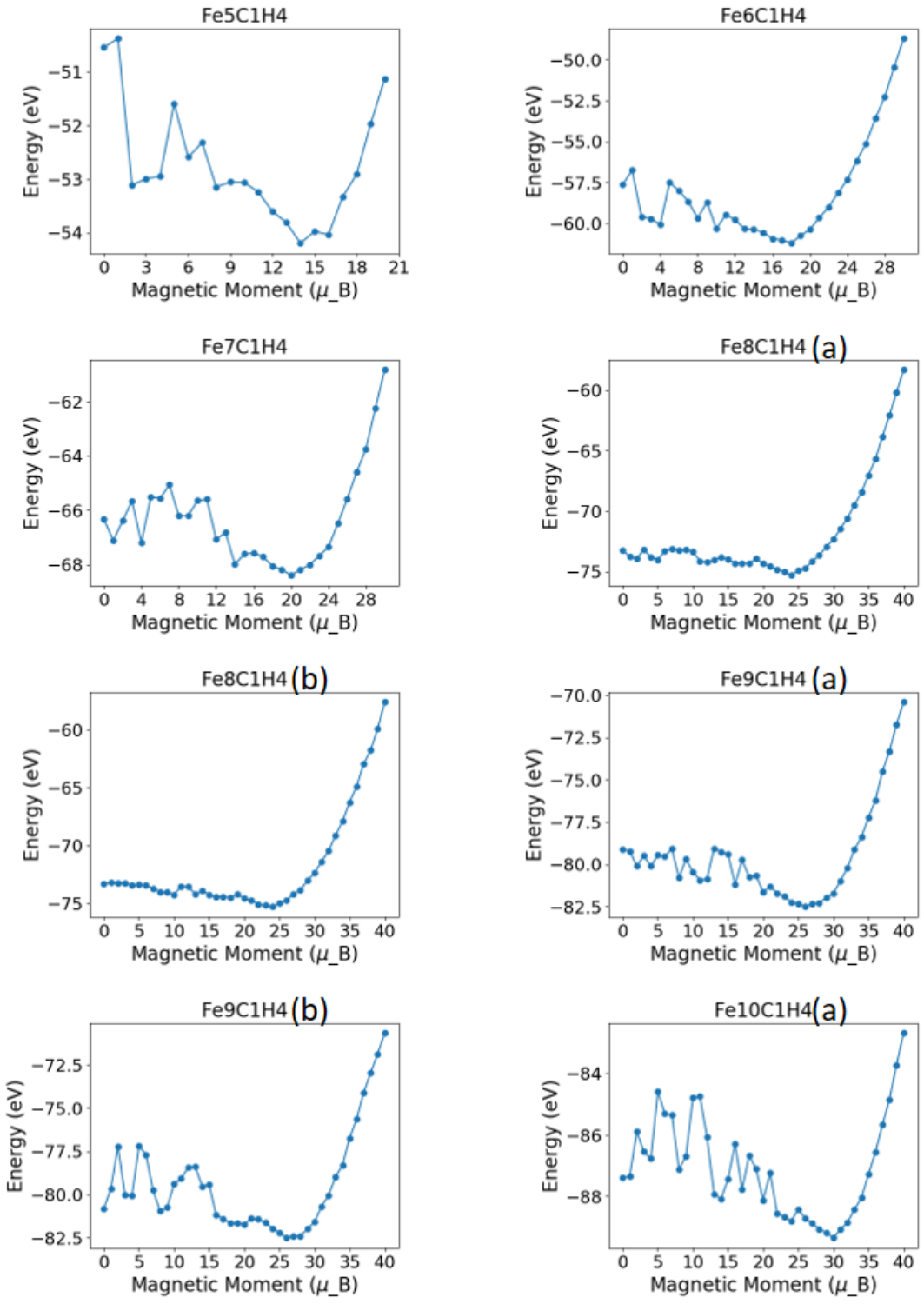

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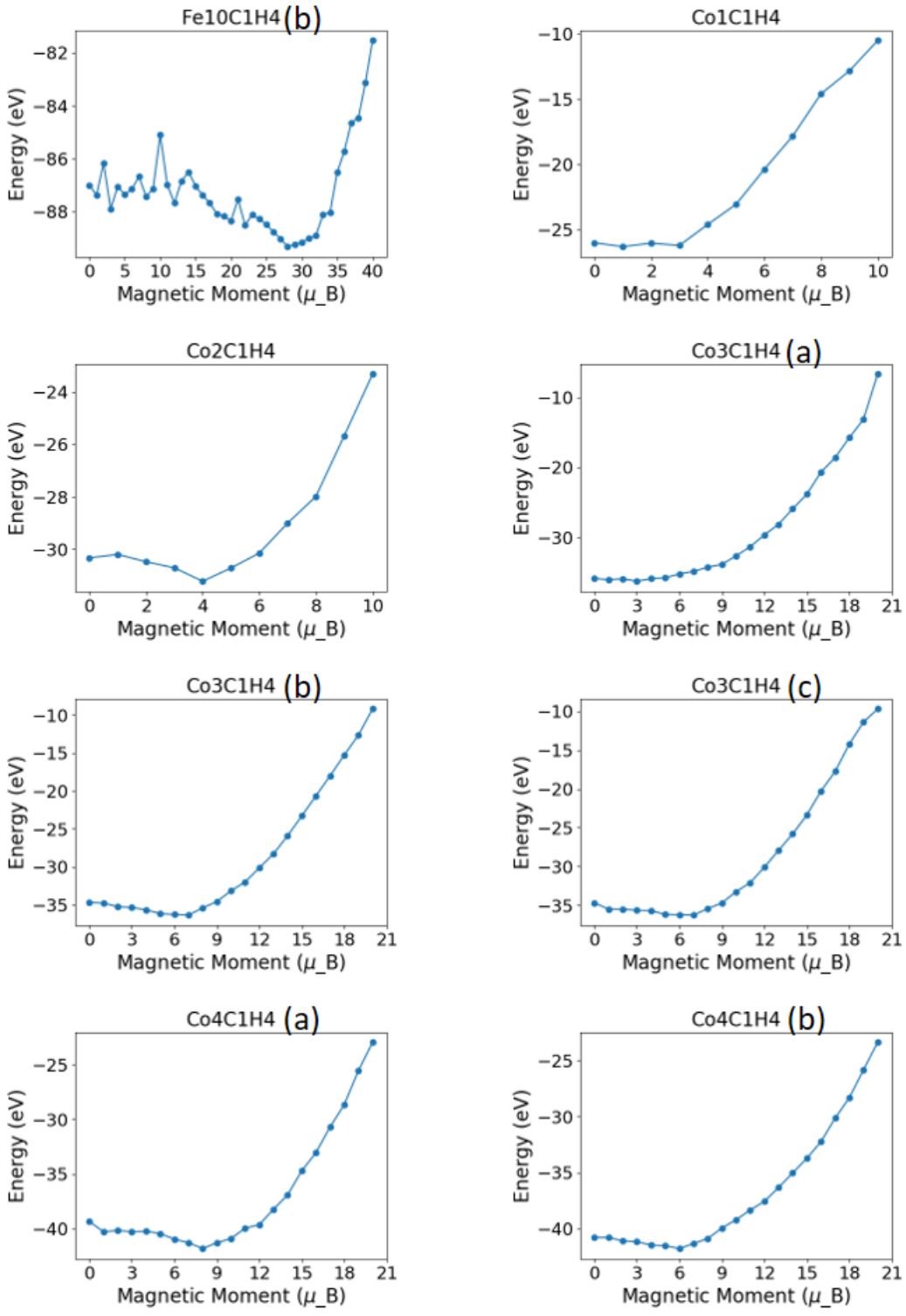

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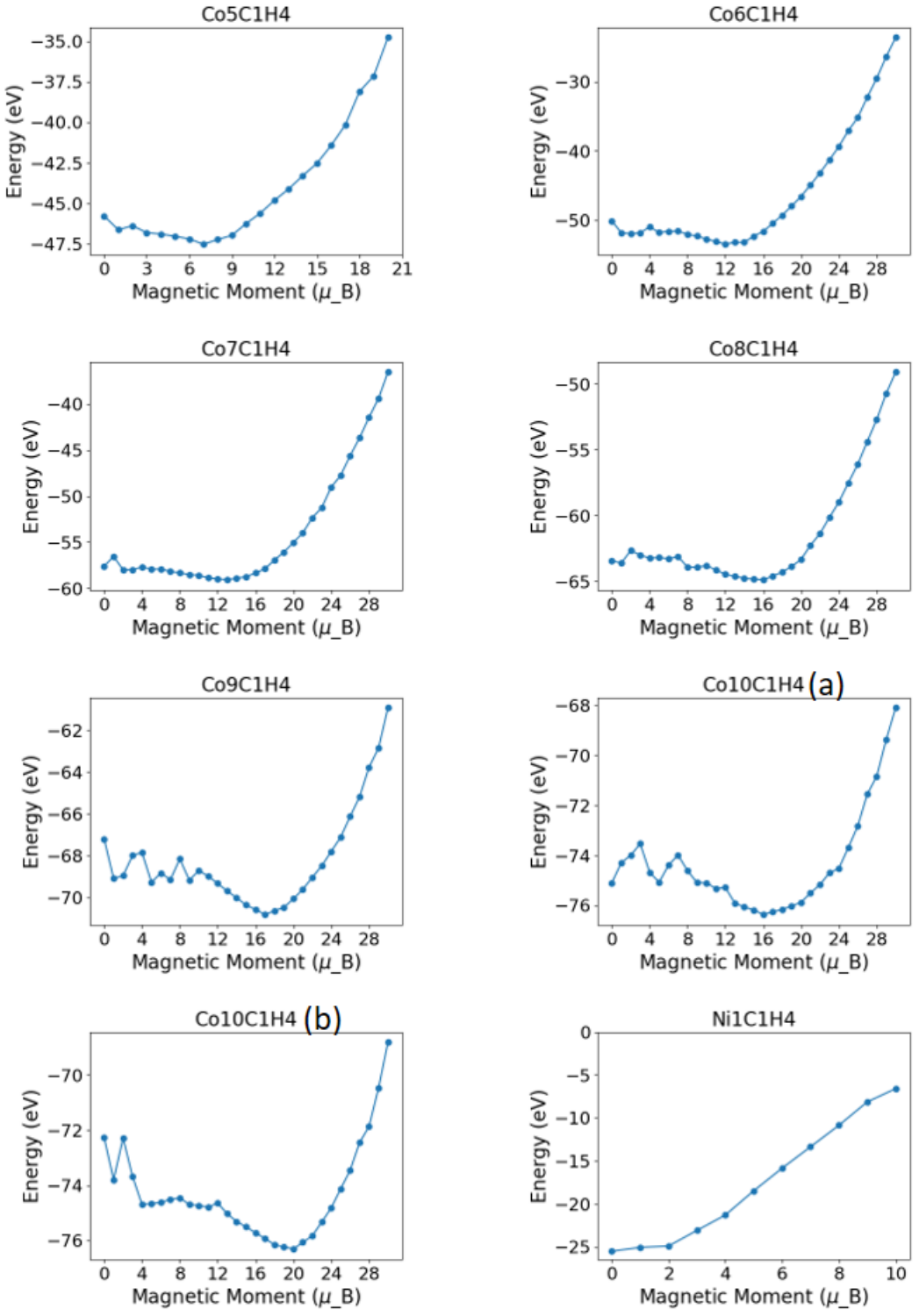

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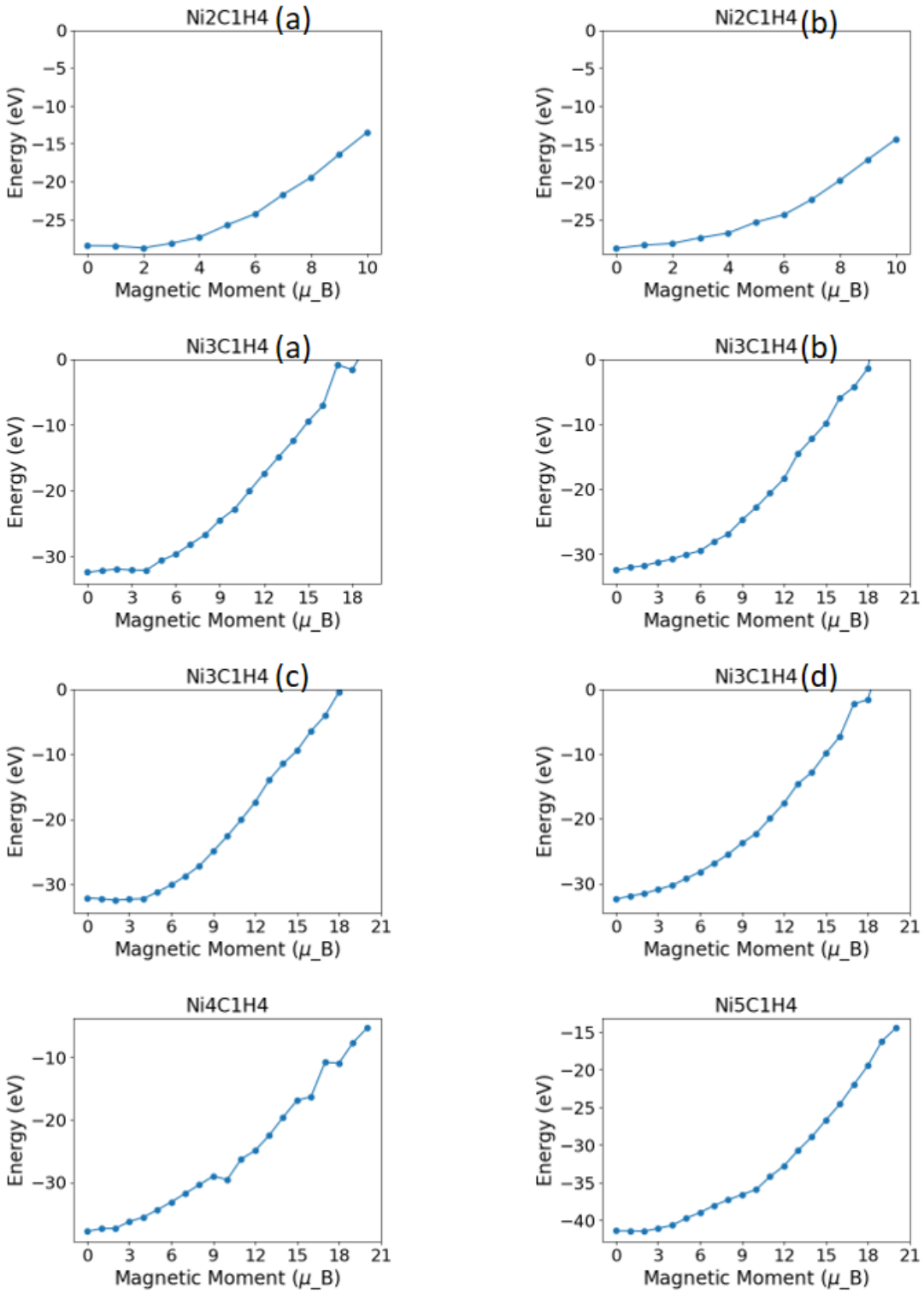

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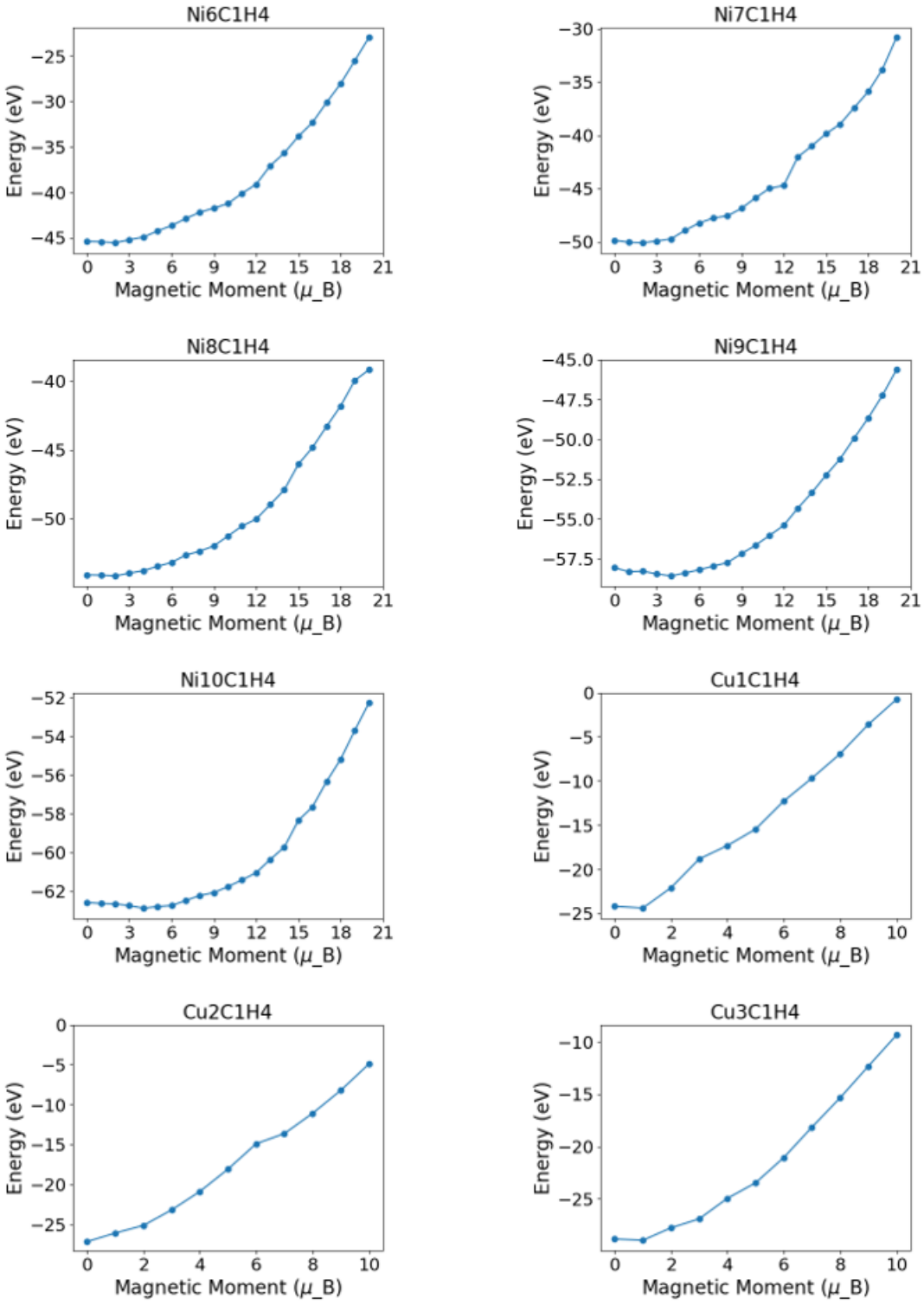

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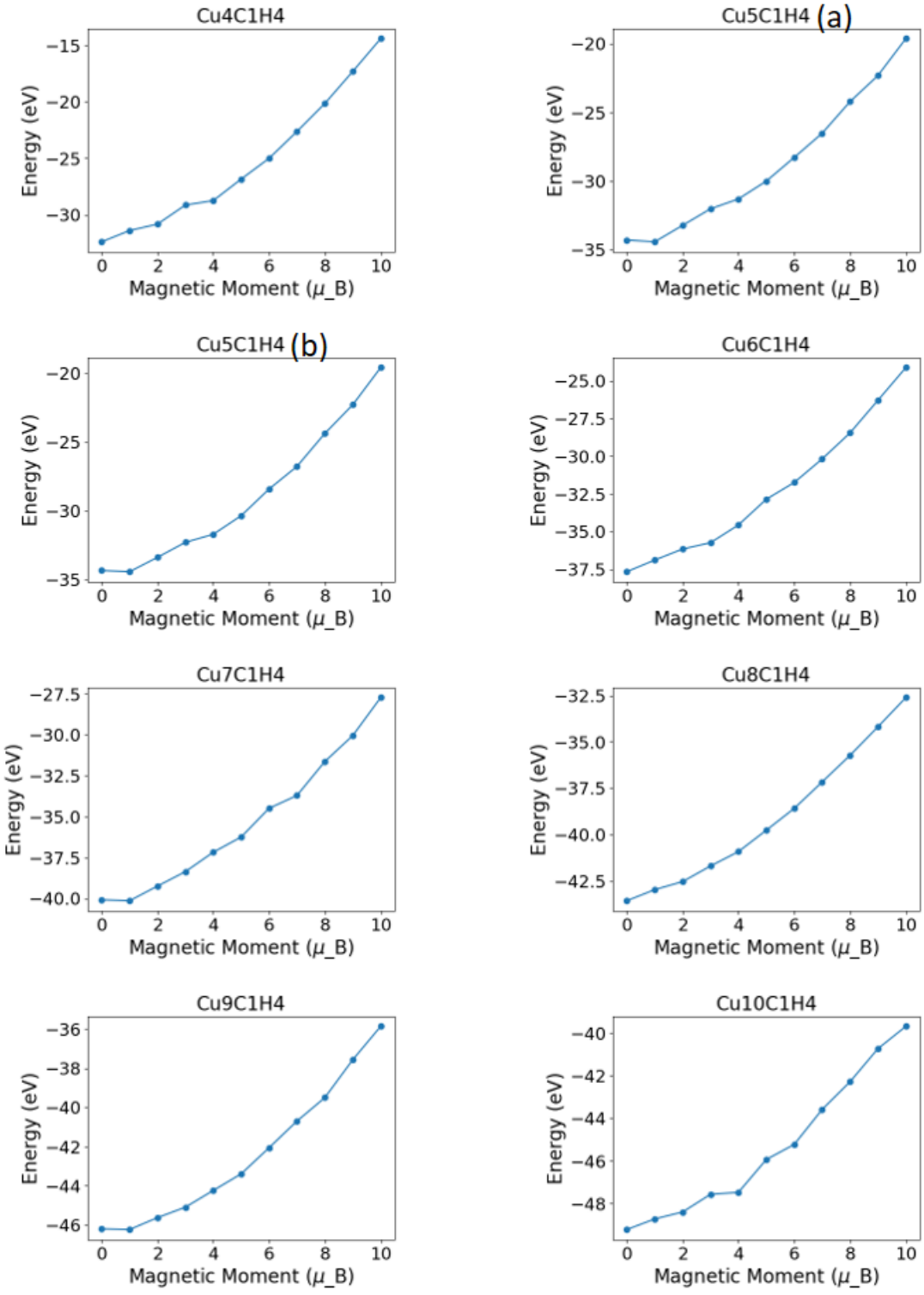

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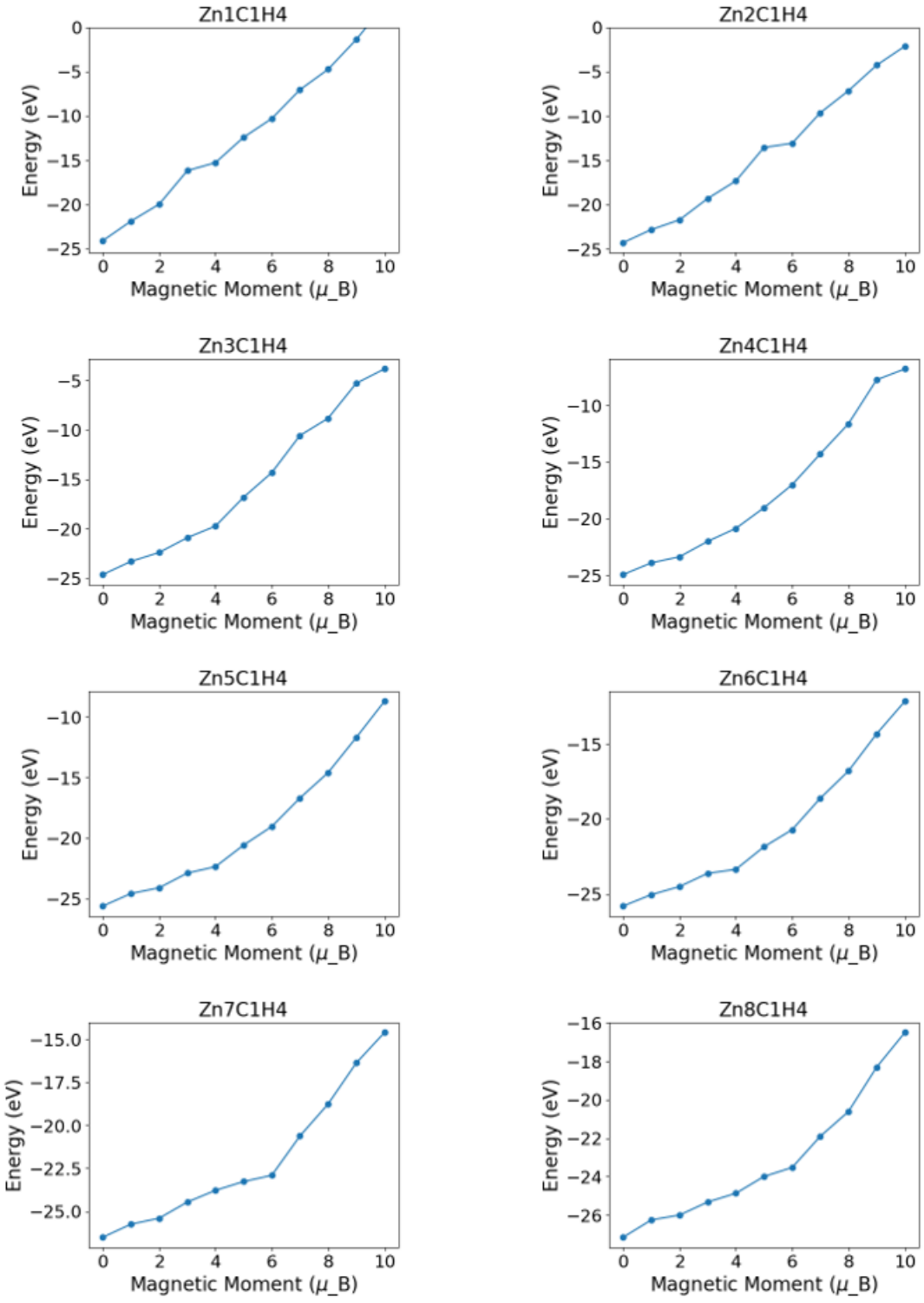

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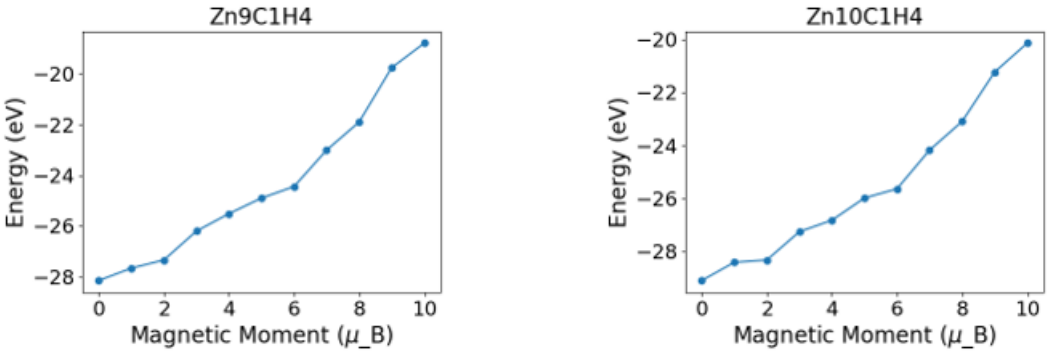


Figure S13. Continued.

S6. Radial distribution function for the M-C bonds

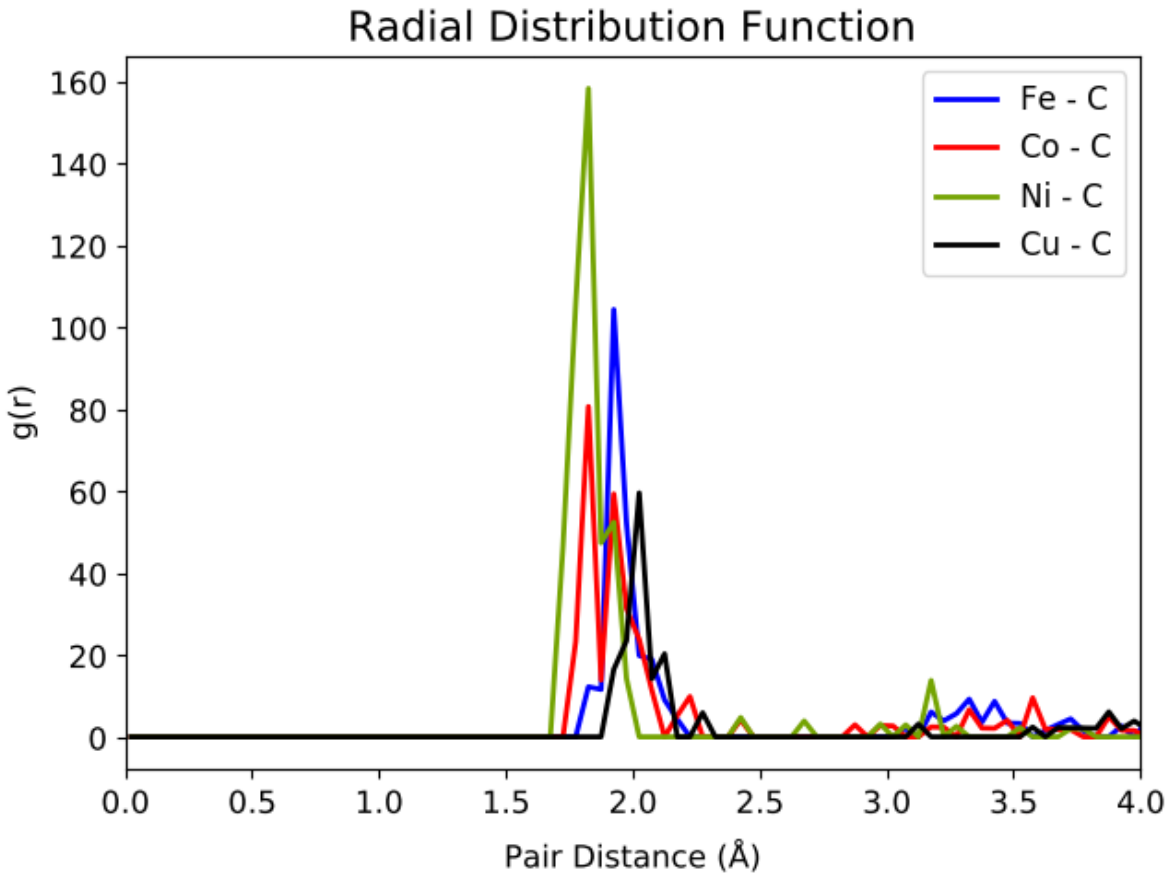


Figure S14. Radial distribution function for the M-C bonds calculated by taking all the structures shown in Figures S3, S5, S7, and S9 into account.

S7. Entropic effects

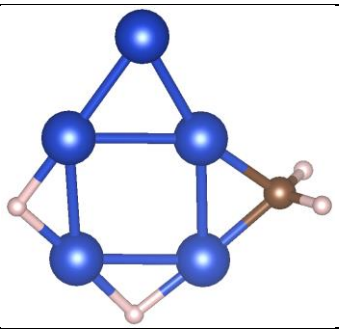
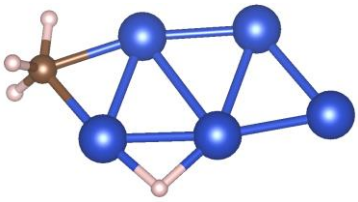
In the main text, we focus on the energy differences between intermediates; however free energy would be a more appropriate quantity to discuss the relative stability of surface adsorbates. While limited computational resources limit us, an estimation of possible error due to the omission of entropic effects would be useful. Thus, we have this section.

We have selected two isomers for Cu_5CH_4 and performed vibrational analysis to estimate how large the entropy difference is. This calculation was carried out by using the DMol^3 module[5,6] implemented in the Materials Studio software.[7] The double numerical plus polarization basis set (DNP)[5] was used in combination with the Perdew-Burke-Ernzerhof (PBE)[8] functional. The Effective Core Potential (ECP) was used to handle the core electrons. The expression for dispersion energy within the method of Tkatchenko and Scheffler was adopted.[9,10] The self-consistent field (SCF) tolerance was set to 1×10^{-6} Ha and SCF convergence was improved by setting smearing to 0.004 Ha. All these calculations were carried out in a spin-unrestricted manner. The direct-inversion iterative-subspace (DIIS) density mixing scheme with charge and spin mixing amplitudes of 0.2 and 0.5, respectively, was used to relax electrons. For geometry optimization, the energy convergence accuracy, maximum force tolerance, and maximum displacement were set to 1×10^{-5} Ha, 2×10^{-3} Ha/Å, and 5×10^{-3} Å, respectively.

Table S4 summarizes the calculated entropy *S* values for the two isomers. We do not see any significant variation between them. To estimate their contribution to the free energy, we also calculated the value of *TS* with *T* being the absolute temperature. The difference in *TS* between the two at 300 K is about 0.03 eV and that at 600 K about 0.05 eV. This implies

that the higher the temperature, the larger the error due to the omission of entropic effects. The tolerance of the energy error to determine the stable adsorbate in our calculation was set to 0.1 eV. At high temperatures, the entropy effect would become non-negligible compared to this value, affecting the determination of which adsorbed species are stable.

Table S4. Optimized geometries of the two isomers for Cu_5CH_4, their entropies, *S*, calculated at 300 K and 600 K, and the temperature-entropy product values of *TS*.

	CH_2-Cu_5	CH_3-Cu_5
Optimized geometry		
S at 300 K (cal/mol·K)	111.6	113.9
TS at 300 K (eV)	1.452	1.481
S at 600 K (cal/mol·K)	139.6	141.5
TS at 600 K (eV)	3.631	3.681

S8. Interaction of the band of metal with an adsorbed level

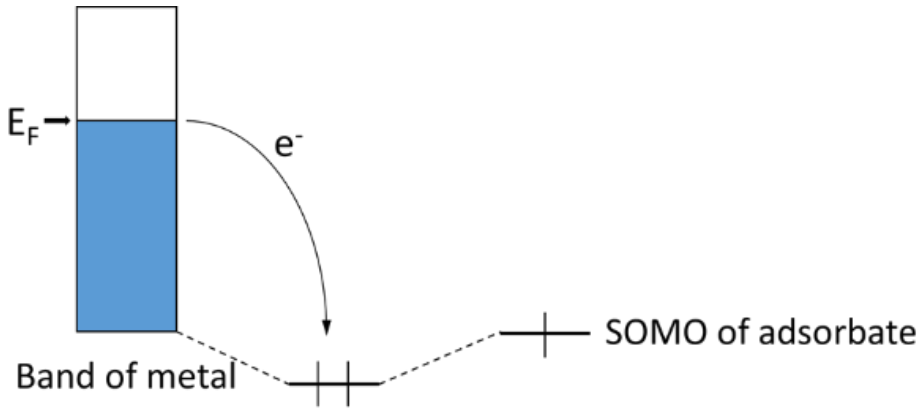


Figure S15. Schematic representation of the interaction of the band of a metal surface (or a metal nanocluster) with the SOMO of an adsorbed species (or a C_1 fragment) upon the formation of an ionic bond between them. The ionic bond is formed when electrons transfer from the Fermi level (E_F) of the metal surface to the SOMO of the adsorbed species and a charge difference is generated between the two. The larger the energy difference between the Fermi level and the SOMO, the greater the stabilization energy of the electrons associated with the charge transfer. This contributes to the strengthening of the ionic bond. This figure was generated on the basis of a paper by Hoffmann and co-workers (see ref. 82 in the main text).

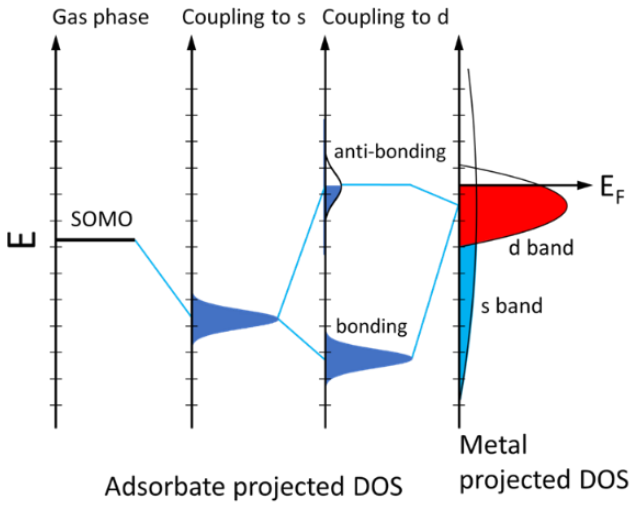


Figure S16. Schematic representation of covalent bonding formation between the s and d bands of a metal surface (or a metal nanocluster) and the SOMO of an adsorbed species (or a C_1 fragment). The bonding and antibonding orbitals (states) formed as a result of the interaction of the SOMO with the d band after its coupling with the s band are shown. It can be seen that the electron occupancy of the d-band affects that of the antibonding orbitals formed between the metal and the adsorbed species. This figure was generated on the basis of a paper by Nørskov and co-workers (see ref. 87 in the main text).

S9. Partial DOS (PDOS) plots for the Fe_{10}, Co_{10}, Ni_{10}, Cu_{10}, and Zn_{10} nanoclusters and those for CH_3 and H

To qualitatively understand the covalency of the M-C and the M-H bonds, we have analyzed the DOS of the metal nanoclusters and the adsorbed species. Figure S17 shows the partial DOS (PDOS) plots for the 3d orbitals of Fe_{10}, Co_{10}, Ni_{10}, Cu_{10}, and Zn_{10}. Figure S18 shows those for the $2p_z$ orbital of C in CH_3 and the 1s orbital of H. The $2p_z$ orbital of CH_3 and the 1s orbital of H in Figure S18 correspond to the SOMO of the adsorbed species.[11] The $2p_z$ orbital of CH_3 may be considered as a representative orbital of the C_1 fragment, responsible for the interaction with the surface. Figure S17 shows that the d-band center gets lower in the order of Fe > Co > Ni > Cu > Zn. From Figures S17 and S18, it is clear that the smallest energy difference between the d-band center and the $2p_z$ orbital of CH_3 is achieved for Ni, consistent with the strong covalent nature of the bonding of Ni to the C_1 fragment. The energy levels of the d-orbitals of Cu and the $2p_z$ orbitals of CH_3 are also close to each other, which could suggest that Cu and C form a strong covalent bond, too. However, the Cu-C bond is weaker than the Ni-C bond because the d-band of Cu is mostly occupied and more electrons could enter the anti-bonding orbitals in the Cu-C bond than in the Ni-C bond. The d-band of Zn is located in the very low energy region and cannot interact with the $2p_z$ orbital of CH_3, so the Zn nanoclusters are unlikely to activate methane.

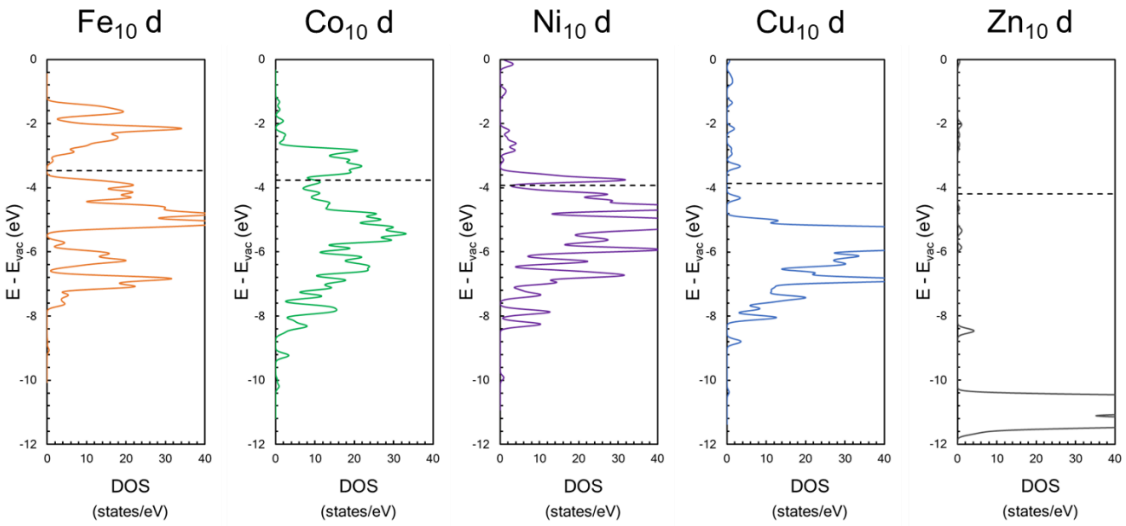


Figure S17. PDOS plots for the 3d orbitals of the Fe_{10}, Co_{10}, Ni_{10}, Cu_{10}, and Zn_{10} nanoclusters without any C_1 fragment adsorbed. These were calculated in a spin-polarized manner. What are shown here are the sum of spin-up and spin-down components. The dashed line denotes the Fermi level.

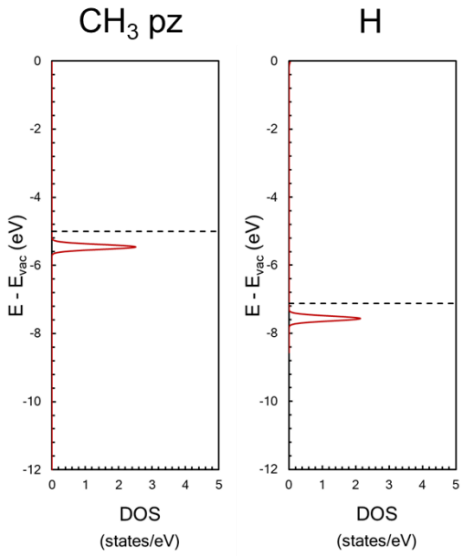


Figure S18. PDOS plot for the $2p_z$ orbital of the C atom in CH_3 and that for the 1s orbital of H, both of which are calculated in the gas phase. The $2p_z$ orbital of CH_3 denotes the 2p orbital oriented perpendicular to the molecular plane of CH_3. These were calculated in a spin-polarized manner. Here only the majority spin component is shown. The dashed line denotes the Fermi level.

From Figures S17 and S18, it is clear that the energy level of the 1s orbital of H lies

lower than that of the $2p_z$ orbital of CH_3, so the energy levels of the 3d orbitals of Cu and the 1s orbital of H are closest to each other. Therefore, although at first glance, the covalent bond of Cu-H appears to be stronger than that of Fe-H, Co-H, and Ni-H, the Cu-H bond is actually weaker because there are more electrons coming from the d-band to the antibonding orbitals of the Cu-H bond. This is consistent with the results of Figure 10.

S10. Coordinates of the optimized CH_4, CH_3, CH_2, CH, C, and H structures

The structures of CH_4, CH_3, CH_2, CH, C, and H were optimized in a large unit cell to make the energy diagram of dehydrogenation reaction of CH_4 to C in the gas phase. The following structural data are provided in the VASP POSCAR format.

```
CH4 structure
C H
   1.00000000000000
    21.4562700000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   21.6984400000000015    0.0000000000000000
     0.0000000000000000    0.0000000000000000   21.7430899999999987
   C    H
     1     4
Selective dynamics
Direct
  0.4830275575139469  0.5072841245619979  0.4950367876004729   T   T   T
  0.5341845863695113  0.5072216269779597  0.4949569551408282   T   T   T
  0.4659695805817945  0.5217967635959050  0.5403243151368750   T   T   T
  0.4659940276698019  0.4606354635970068  0.4848834235761904   T   T   T
  0.4659849444809371  0.5392766062781332  0.4597231957656289   T   T   T


CH3 structure
C H
   1.00000000000000
    21.5536011200900006    0.0000000000000000    0.0000000000000000
     0.0000000000000000   21.8217090131000013    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.9726868182040000
   C    H
     1     3
Selective dynamics
Direct
  0.4894572121754927  0.5044072779103029  0.4999999933424211   T   T   T
  0.5398261975368568  0.5082598676482538  0.5000000029929614   T   T   T
  0.4678146579406988  0.4593515456565218  0.5000000016119792   T   T   T
  0.4608864890259531  0.5454930093829142  0.5000000020526381   T   T   T


CH2 structure
C H
   1.00000000000000
    21.1877203206820006    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.1267130133349994    0.0000000000000000
     0.0000000000000000    0.0000000000000000   21.9026728017060002
   C    H
     1     2
Selective dynamics
Direct
  0.4895060557737845  0.5070236396853810  0.4934310233762152   T   T   T
  0.5295232672907159  0.5001400821737826  0.4631021233074973   T   T   T
```

```
  0.4792215231895020  0.4946717372308396  0.5407011425382873   T   T   T


CH structure
C H
   1.00000000000000
    20.5567995799709990    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.1002982650109985    0.0000000000000000
     0.0000000000000000    0.0000000000000000   21.0119128590799988
   C    H
     1     1
Selective dynamics
Direct
  0.5132927384661506  0.5024672534884344  0.4763775306250178   T   T   T
  0.4867072615338493  0.4975327465115657  0.5236224693749824   T   T   T


C structure
C
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   C
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


H structure
H
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   H
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T
```

S11. Coordinates of the optimized cluster structures

The followings are the atomic coordinates of the nanocluster structures shown in Figures S3, S4, S5, S6, S7, S8, S9, S10, S11, and S12. The following structural data are provided in the VASP POSCAR format.

```
Fe1CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    17.4776309000000012    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9679985000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9487696999999997
   Fe   C    H
     1     1     4
Direct
  0.5197184125336924  0.4268763365206686  0.4850288060301264
  0.4932140125521104  0.5228798444986187  0.5341222572849615
  0.5304760326638894  0.5671306812998734  0.5117126817360745
  0.5368983696785953  0.3866963600765566  0.4061312089378820
  0.4332181018926362  0.5369590323618810  0.5208195219282159
  0.4996950706790800  0.5212677452424023  0.5953255240827386


Fe2CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    17.9497299000000012    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4035782999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1794009000000010
   Fe   C    H
     2     1     4
Direct
  0.5077437781273693  0.3622984453441049  0.4791609165138992
  0.5282119123463906  0.4711437046485939  0.4697939642391581
  0.4528593725087765  0.5459114823131602  0.4914798309202512
  0.4041524594472916  0.5401708846786872  0.4549246572097471
  0.4325543941233413  0.5436328501530420  0.5487359380117869
  0.5881915523956324  0.3957030911959154  0.4512185871833169
  0.4775465310511868  0.6002095416665008  0.4819061059218345


Fe3CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    18.7241472999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.7439289000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.5992424000000014
   Fe   C    H
     3     1     4
Direct
  0.5151054451274060  0.4826864241899159  0.5329691022205666
```

```
  0.4611724684542249  0.5250199086823863  0.4093136925292981
  0.4250603983479170  0.5591602756621944  0.5160627162188410
  0.5391797216973615  0.4575260979254334  0.4339842092397673
  0.4641254609868142  0.5339119284523867  0.5954329653466827
  0.5295745605883810  0.3975981419024800  0.4207954639357140
  0.5925941338419707  0.4762524423791190  0.4162529454542591
  0.3886678109559197  0.5822447808060789  0.4347489050548716


Fe4CH4(a) shown in Figure S3
1:P1(1)
   1.00000000000000
    19.3046213999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.3164985999999992    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.3797543999999995
   Fe   C    H
     4     1     4
Direct
  0.4328417642954854  0.4918501477807128  0.4389858716113859
  0.5646051289102639  0.4553993801023922  0.4588685671488150
  0.4829542348859782  0.4600238138741498  0.5391457253201061
  0.5222604912483257  0.5656312833013224  0.4674228270397976
  0.4350037761282202  0.5595058102071481  0.5196749866884935
  0.4274580635029575  0.5298687430309318  0.5726256176065482
  0.6017803329783132  0.5352105186497493  0.4323805176834684
  0.3951476219130902  0.6021262346205805  0.5247213557897128
  0.5393485861373606  0.3920540684330204  0.5220345311116777


Fe4CH4(b) shown in Figure S3
50:P1(1)
   1.00000000000000
    18.1046881000000006    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.0799497000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.2027740999999992
   Fe   C    H
     4     1     4
Direct
  0.4422450585486929  0.5548825765497225  0.5426191195076672
  0.5273017149002504  0.5651896703410001  0.4522330411200307
  0.5629524115797171  0.4710494071922025  0.5282636822952361
  0.4457657177161267  0.4373446670281834  0.4926251828315949
  0.5285558253871436  0.4552022987071315  0.4292821584683392
  0.3851889351122396  0.4773067445339004  0.5574274656867144
  0.4748167054636719  0.6358743866650469  0.4972622850160366
  0.6072685563825809  0.5538498244675674  0.5023676207266597
  0.5561550749095864  0.4217204245152533  0.3855294443477235


Fe4CH4(c) shown in Figure S3
34:P1(1)
   1.00000000000000
    19.4088436999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.9084864000000010    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.2042871999999996
```

```
   Fe   C    H
     4     1     4
Direct
  0.5427107772040282  0.4723707557282358  0.5091546583880320
  0.5982817365336430  0.5472819634176328  0.4330908355445972
  0.5743337293764602  0.5851497916377467  0.5550962745143744
  0.4821571331010787  0.5686268059380175  0.4759706089234266
  0.4299789138519413  0.4806249530116420  0.5171486592172714
  0.4482217941035333  0.4280913659259029  0.5393866448771849
  0.6188761544913579  0.4622991722965458  0.4611267562666241
  0.3991860088931022  0.5021114738865540  0.5655695005074587
  0.3955937524448463  0.4667337181577277  0.4683260617610308


Fe5CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    17.6268324000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.5230791000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.1963039000000002
   Fe   C    H
     5     1     4
Direct
  0.4898217817994546  0.5210481691912605  0.5813632486228111
  0.5536270969677506  0.6009994288546752  0.5156473472996673
  0.4555155074121887  0.4321343387550410  0.5072693803225951
  0.5556348063330331  0.4865759509483755  0.4601680838506129
  0.4360007153207205  0.5622860843248475  0.4667315003060032
  0.4572905967919976  0.4738328113342520  0.4190031024010883
  0.4506994545268087  0.4313633432328224  0.5915308853975771
  0.4442524206160814  0.4629783386667575  0.3665518600710705
  0.5244427289940686  0.6117588476361600  0.5942177466033643
  0.6123348912379007  0.5672126870558100  0.4564168451252104


Fe6CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    17.9045692000000010    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.5705807000000007    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.2792312000000017
   Fe   C    H
     6     1     4
Direct
  0.4243043295512639  0.4392175563324273  0.5363608593949729
  0.4838124388175001  0.5339488467949158  0.5869972255692207
  0.4496405466940916  0.5469280489805001  0.4733484537547691
  0.4959859483789497  0.4205934655631489  0.4325860437045908
  0.5678314783696736  0.4546338933648911  0.5306520861484104
  0.5699516509744578  0.5202487447943863  0.4293126021467387
  0.5538797284466638  0.5656702397857456  0.5178394280537002
  0.4110501045717372  0.4790008175102003  0.6154249081551320
  0.5582098759388002  0.4495322166489488  0.3710455973006118
  0.5891607179326129  0.6132954475118301  0.5286144437492336
  0.4237631803242529  0.3783107227129947  0.4712983520226178
```

```
Fe7CH4 shown in Figure S3
1:P1(1)
   1.00000000000000
    18.2510635999999984    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.7138124999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.6184490999999994
   Fe   C    H
     7     1     4
Direct
  0.4202642814306755  0.5483727734713736  0.4999899611464107
  0.5661541177998248  0.5665203015385392  0.5399241834875949
  0.4514097554427511  0.4258583799775671  0.4833387285969308
  0.5902682554603710  0.4501303479444131  0.4851924491132621
  0.5186991239530836  0.5596506697957012  0.4272995913577217
  0.5127273337453390  0.4590147732496994  0.5846786745780819
  0.5171944529812094  0.4403861428648262  0.3797753682226037
  0.4294626289035665  0.4996136797782436  0.4049602537016857
  0.3853913806877821  0.5041423972033886  0.3645996775486290
  0.5646307647048155  0.5271221454374669  0.6225542254769389
  0.5827983607379433  0.6175300993094214  0.4650402804759715
  0.4657595441526424  0.3801282894293684  0.5628966062941708


Fe8CH4(a) shown in Figure S3
1:P1(1)
   1.00000000000000
    18.4992252000000015    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4462770999999996    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.1554651000000007
   Fe   C    H
     8     1     4
Direct
  0.6067923192590982  0.4907845551251289  0.5398140524677343
  0.4822813946046767  0.5460911094434471  0.5182577877689173
  0.5512540978029141  0.3807002709045879  0.5709737984946091
  0.4273876923956856  0.5414544147473953  0.6239841594982197
  0.5574816968715250  0.5884087685211851  0.6021103043375283
  0.4329458500314511  0.4276925586527447  0.5694132454662016
  0.5295009245741034  0.4782796557833121  0.6470867430776741
  0.5120578669402768  0.4370712301158524  0.4697463630682184
  0.4757955021505423  0.5247200758967121  0.4088753752118708
  0.5043971348419382  0.5758307926027838  0.3968251177212249
  0.4176417146367797  0.5312383402201428  0.3978771907122035
  0.4967588814058701  0.4860479545499392  0.3688522240996653
  0.5442349244851329  0.3528302734367698  0.4864936380759335


Fe8CH4(b) shown in Figure S3
4:P1(1)
   1.00000000000000
    19.8628773999999986    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.5196725999999998    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.8087273999999987
```

```
   Fe   C    H
     8     1     4
Direct
  0.6072191098787957  0.4916599974419618  0.5417698648051289
  0.4446193150507171  0.5614614384995705  0.4920782268774927
  0.4962834771637520  0.4621957468733677  0.5540477366125914
  0.4598971567068295  0.4820042701607073  0.4022292998635109
  0.4581972846534557  0.3831726301646516  0.4717628719726883
  0.5483398605018238  0.5922225154932425  0.5380897821012035
  0.5588315210438126  0.5396660355661500  0.4287867921207217
  0.5670047984793450  0.4228534112639148  0.4464345264937043
  0.4109303074679064  0.4683976050273406  0.4939379702072930
  0.6321583355878113  0.5761452706808720  0.5316266873475883
  0.3569327034558053  0.4603932868019510  0.5056344463399753
  0.4744399048141647  0.6362620841568369  0.5269333469453558
  0.5680262251957913  0.4460957078694439  0.6044284483127588


Fe9CH4(a) shown in Figure S3
1:P1(1)
   1.00000000000000
    19.1853352000000008    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.5578967000000006    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.9126775000000009
   Fe   C    H
     9     1     4
Direct
  0.4580418492716887  0.4537606902488941  0.5799628130463483
  0.4619544449668689  0.4564520756362540  0.3971471531880955
  0.4023774556749518  0.5499441263972671  0.5517761421269942
  0.4538959063133479  0.3811364340007026  0.4914836857177710
  0.5208702440487148  0.5525775184939589  0.5271698832323247
  0.4322530838664583  0.5590257676334286  0.4341717716628906
  0.5528201495912011  0.4464704060517501  0.4886366688321329
  0.5476192311759042  0.5334548252193536  0.4156138155532597
  0.3738199598974351  0.4633918604001202  0.4845155951823604
  0.5527408674213998  0.4801784924674430  0.5782530168508083
  0.5555401903723173  0.4469881634637520  0.3972485215810731
  0.5089826044146047  0.6052799854968213  0.4540810875159272
  0.3801953344507836  0.4971293844094961  0.3966303209720889
  0.5864886785343144  0.4751302700807579  0.6230195245379327


Fe9CH4(b) shown in Figure S3
2:P1(1)
   1.00000000000000
    18.7236657999999991    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.9035042000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.0580770999999984
   Fe   C    H
     9     1     4
Direct
  0.4643284285495763  0.4316850652969608  0.4310436254095994
  0.4763454970404054  0.5618833318020933  0.3959813164555958
  0.3909765345110003  0.5216709835074529  0.4773555613304497
```

```
  0.5454551722112500  0.5342364626071598  0.5744739905951763
  0.4237408843819047  0.5500977361367351  0.5912717590427772
  0.4582658796401204  0.4366243997152864  0.5529207630745343
  0.5802766691678275  0.5700992001389994  0.4612867050412741
  0.4754540881304181  0.6207843483007947  0.5031473281956940
  0.5681770921508055  0.4404200225322872  0.4963483035842383
  0.5458748486784282  0.4891436892072322  0.4107427462197112
  0.5458104399819860  0.4413166182470294  0.5907400049739747
  0.5626855083537087  0.6211070197295865  0.5419854833114145
  0.3869144428200706  0.4282822065115003  0.4907640405293846
  0.4036145143824976  0.6081889162668788  0.4419883722361709


Fe10CH4(a) shown in Figure S3
1:P1(1)
   1.00000000000000
    19.5215341000000002    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.2506552000000006    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.0800531000000007
   Fe   C    H
    10     1     4
Direct
  0.5415861687525818  0.4279103928080357  0.6037607594289566
  0.4780564168997142  0.4699227504025373  0.4311020226199481
  0.5009319015640727  0.5480541018916455  0.6042787117120414
  0.4924673874576561  0.5835179631134899  0.4894552861590504
  0.4264573029687752  0.4522891709118684  0.6330578817583856
  0.5937408449099706  0.4461672930318120  0.4078979438869788
  0.4384894935780173  0.3916168520763583  0.5268165436998234
  0.5516838160161834  0.3713883020703763  0.4947939167192033
  0.4012966750385795  0.5124191436448635  0.5245703747070067
  0.5875603218738854  0.5077378089902759  0.5153813841617220
  0.5561409875314532  0.5425053303014077  0.4198543045759996
  0.4334762278135044  0.6000532623887723  0.5594717425544643
  0.5753804233665575  0.5806137508342062  0.3789927518864449
  0.3922362054869713  0.4402517791764506  0.4608297345331278
  0.5867958267420779  0.5112420983579044  0.6131266415968469


Fe10CH4(b) shown in Figure S3
4:P1(1)
   1.00000000000000
    19.7221627999999995    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.3298564000000006    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.2565474999999999
   Fe   C    H
    10     1     4
Direct
  0.4877680520002526  0.3833373060567354  0.5373546361827833
  0.4977292212215394  0.5940807889135392  0.5454792857634402
  0.5126211081260412  0.4906185518341515  0.6103179453268880
  0.4137435360905721  0.4848152245491729  0.5466283418629067
  0.6042206766563843  0.5340013093016828  0.5366416869831830
  0.4386892701272979  0.4301712441781225  0.4376701986974862
  0.5525185435000133  0.5731641335373331  0.4355117764171253
```

```
  0.4325363628691390  0.5608428412706246  0.4489266382585999
  0.5627958907611531  0.4419661380138719  0.4577784634682617
  0.5980228298630775  0.4107032562581960  0.5737172638854124
  0.4976492920717138  0.4989637215890952  0.4047489904894527
  0.4981841912713158  0.3625110192557374  0.4479183071758928
  0.4877439924039711  0.6359000574711602  0.4486262526445229
  0.6234459815369229  0.5202063753979841  0.4467461366859847
  0.3719610515006073  0.4911080323726021  0.4543640761580620


Fe1 shown in Figure S4
Fe
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Fe
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


Fe2 shown in Figure S4
Fe
   1.00000000000000
    22.6700000000000017    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Fe
     2
Selective dynamics
Direct
  0.4563253380608873  0.4999568518257836  0.4998277539134800   T   T   T
  0.5436746619391127  0.5000431481742164  0.5001722460865200   T   T   T


Fe3 shown in Figure S4
1:P1(1)
   1.00000000000000
    15.0234114000000005    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.1949587000000008    0.0000000000000000
     0.0000000000000000    0.0000000000000000   15.9191161000000001
   Fe
     3
Direct
  0.4999392427734068  0.5044105079505578  0.4317321956077317
  0.5006523086177964  0.4316191313933064  0.5527144222948704
  0.4994084486088038  0.5639803606561318  0.5444233820973957


Fe4 shown in Figure S4
1:P1(1)
   1.00000000000000
    16.3874591999999986    0.0000000000000000    0.0000000000000000
```

```
     0.0000000000000000   16.6020766000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.5972031000000015
   Fe
     4
Direct
  0.4590461270786228  0.4347125141349954  0.4333424614481429
  0.4430528602221997  0.4476637455338058  0.5668101019462978
  0.5648746359793705  0.5007964914294610  0.4878454076906222
  0.4483563767198043  0.5685772489017350  0.5074820289149302


Fe5 shown in Figure S4
1:P1(1)
   1.00000000000000
    16.9951049000000012    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.9951049000000012    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.2144350000000017
   Fe
     5
Direct
  0.4438993039093918  0.5562267982877347  0.4467968184479484
  0.5562732244531092  0.4442278537084259  0.4458900488479987
  0.4239291482667611  0.4235531040384584  0.4673899999085472
  0.5762294542819237  0.5762697018214058  0.4676326946067299
  0.4996788690888174  0.4997225421439752  0.5599404381887766


Fe6 shown in Figure S4
1:P1(1)
   1.00000000000000
    18.0784066999999986    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.1382690000000011    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1790225999999997
   Fe
     6
Direct
  0.5046250762269322  0.4427731254946591  0.5638208204600688
  0.5925908479103759  0.5136777737074072  0.4774383683292073
  0.3964389805559853  0.4767389755748631  0.4802139891418706
  0.5059705384158475  0.4241287043686670  0.4383618823805968
  0.4844672357042052  0.5477338995894981  0.3935297444537745
  0.4829573211866612  0.5663775212649178  0.5189851952344794


Fe7 shown in Figure S4
2:P1(1)
   1.00000000000000
    18.0546867999999989    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.2272897000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1342485999999994
   Fe
     7
Direct
  0.4831905100274860  0.4725350501019600  0.5828102724493271
  0.5335313748177521  0.5128960148698682  0.4425585331282454
```

```
  0.4056225516298095  0.4657848289151180  0.4805805681296800
  0.5106631249220007  0.3954858170104528  0.4832593621776374
  0.6062783870935603  0.4721168922115911  0.5392747685520006
  0.5487798533950157  0.5813964998889072  0.5657297225724442
  0.4438941981143738  0.5826248970021058  0.4977667729906649


Fe8 shown in Figure S4
1:P1(1)
   1.00000000000000
    18.2110504999999989    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9905131999999988    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.8514532999999993
   Fe
     8
Direct
  0.5138341244640824  0.5285975300954999  0.5846994482691350
  0.4711112740444252  0.5941596893201538  0.4833851783081875
  0.4688090708826803  0.4120570120183601  0.5886168195899693
  0.5836121944755266  0.5348504630380521  0.4599581384976838
  0.6057654641544408  0.4509135188982942  0.5530837166155590
  0.5188027209698642  0.4082025301600281  0.4726065230018008
  0.4046941457152201  0.4838615369320223  0.5056398431078932
  0.4755210052937672  0.5010077195375933  0.3961703326097694


Fe9 shown in Figure S4
1:P1(1)
   1.00000000000000
    17.9628404000000010    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.2915446999999993    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.3033083999999988
   Fe
     9
Direct
  0.5248727734626033  0.5785931836196273  0.5403144571587802
  0.5983588502094471  0.4853350877846941  0.5662989944856361
  0.3978448906225671  0.4157211243965340  0.5305265259687021
  0.5746504705273240  0.4961444986752602  0.4378309528562536
  0.4732848658065140  0.5668956762666294  0.4196133176889151
  0.4441941055106468  0.4507598288033824  0.4143445833024035
  0.5255290873487327  0.4012378718516414  0.5007074877690892
  0.4732164089095243  0.4881577516214045  0.6086075414215982
  0.3941185476026481  0.5331249769808212  0.5168961393486200


Fe10 shown in Figure S4
1:P1(1)
   1.00000000000000
    18.2963149999999999    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.2963149999999999    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.8649127999999990
   Fe
    10
Direct
```

```
  0.4998435552995160  0.5002897248642486  0.6250174276361055
  0.5000381925962489  0.4998087057620422  0.3749858809550443
  0.5374914308220732  0.5898640129004402  0.4515944059378846
  0.4626332832677125  0.4099779137015770  0.4518445575419329
  0.4100894455701899  0.5373795379093346  0.4516677121641013
  0.5899781351259501  0.4624286485390397  0.4516583753393020
  0.5373576679534334  0.4102003685517698  0.5484876970360960
  0.4626390212685411  0.5900261279494766  0.5482040318694960
  0.5900267717114142  0.5373725754647206  0.5481866794035546
  0.4098924963849172  0.4626423843573474  0.5483432321164797


Co1CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    16.5025842999999988    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.9364950000000007    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.0038142999999984
   Co   C    H
     1     1     4
Direct
  0.4895678696725292  0.4546788726996168  0.5568719535611089
  0.5082355391569479  0.4885976478100471  0.4604598883114868
  0.5310966195602497  0.4325109926940068  0.4353988951285475
  0.4708478865146715  0.5266769565865249  0.5988498288391895
  0.5546037898052837  0.5350368751079131  0.4555805548510413
  0.4518682952903241  0.5072886551018890  0.4324188793086257


Co2CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    18.1531789000000003    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.5652693000000006    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.1684350999999999
   Co   C    H
     2     1     4
Direct
  0.5887169447903881  0.4525748856867087  0.4903377834236396
  0.5256242687169663  0.5283138512710980  0.5418524592501706
  0.4985354160150976  0.4761914211920493  0.4406073199994227
  0.4586640958309280  0.5180911016895713  0.4658943891456649
  0.5551635683759438  0.5554279564692705  0.6119486210379499
  0.5115437531009849  0.5015597264713347  0.3894062913718674
  0.4656719531696863  0.4239310572199637  0.4337031357712862


Co3CH4(a) shown in Figure S5
1:P1(1)
   1.00000000000000
    18.3075380000000010    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.3496082999999999    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.4389251999999999
   Co   C    H
     3     1     4
```

```
Direct
  0.5191538744969846  0.5298258759342510  0.5542627735294212
  0.5566369601454197  0.5539859111302984  0.4395977416256308
  0.4357805015011197  0.5445240332819821  0.4645964406794457
  0.5062698127217082  0.4768183534897467  0.4730410509215339
  0.5084109669957323  0.4179407587229298  0.4613545898337003
  0.5950004016603690  0.5665812240949946  0.5204630736557676
  0.4821001504237337  0.5863922153191791  0.3990488882755338
  0.4314473320549297  0.5535616280266187  0.5541954414789664


Co3CH4(b) shown in Figure S5
39:P1(1)
   1.00000000000000
    18.9643611000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.7814608999999990    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.2974883999999989
   Co   C    H
     3     1     4
Direct
  0.5368185323865254  0.5249870986046411  0.5533458588273956
  0.5426789443929746  0.5896867444276770  0.4596690200128795
  0.4438995991273170  0.5360340980772947  0.4846574006318534
  0.4539942859007493  0.4658825536233274  0.5567190896858187
  0.4622892082746101  0.4111736857090730  0.5373992373034842
  0.6042522927969382  0.5794054763899299  0.5186100561834043
  0.4675219042401266  0.5954002879121707  0.4175579697268272
  0.4195652328807554  0.4668400552558928  0.6026913676283259


Co3CH4(c) shown in Figure S5
29:P1(1)
   1.00000000000000
    17.5310337999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9129648000000010    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1506350000000012
   Co   C    H
     3     1     4
Direct
  0.4805372147617221  0.5330493682591153  0.5391027669382970
  0.5496564189280121  0.4325468316323779  0.5425290102326117
  0.5121178138590458  0.4760571745152555  0.6427251160634875
  0.5134447964638157  0.4904958995629088  0.4408205172150680
  0.4581689261167115  0.5554650370616045  0.6238893783415628
  0.5673356472955187  0.4655346753127519  0.4205675806743151
  0.5150254416790374  0.5478459368640368  0.4175158571871730
  0.4659837408961280  0.4600250767919534  0.4146897733474788


Co4CH4(a) shown in Figure S5
1:P1(1)
   1.00000000000000
    17.4070722999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.4740867000000009    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.7201442000000000
```

```
   Co   C    H
     4     1     4
Direct
  0.5335779103926727  0.4309351905647095  0.5128616465379501
  0.6035278920358795  0.4646117689522115  0.6104587658305045
  0.5963292815639077  0.5828660063933105  0.6473073374751115
  0.5250325673596681  0.5499989946084655  0.5510129961934278
  0.4638997095383634  0.5088737516872410  0.4578925313339240
  0.4044951858144444  0.5237051883330198  0.4740755098555408
  0.4576546729157009  0.4530825706938651  0.4271671164193222
  0.5933226034842997  0.3790911243439717  0.5633585246891056
  0.4844801768950646  0.5490254044232019  0.4142955716651019


Co4CH4(b) shown in Figure S5
8:P1(1)
   1.00000000000000
    18.2767705000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.2767705000000014    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.4654892000000004
   Co   C    H
     4     1     4
Direct
  0.5559618373198404  0.5690215129711645  0.4950463880333496
  0.4440772020636038  0.4309614554919612  0.4969603314101688
  0.4309291679956887  0.5559767922457940  0.4963890102570226
  0.5690719727456405  0.4440411269562029  0.4961251205047722
  0.5003060055268858  0.5003139425934794  0.5452761110757527
  0.6273319445483778  0.5132418544237041  0.4820807903905658
  0.3724328102720108  0.4866501934503856  0.4850413801550568
  0.4865962718847647  0.6274081926602005  0.4827210466325966
  0.5132927876431947  0.3723749292071044  0.4845398215407210


Co5CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    19.4177284999999991    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.6114695999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.5655369999999991
   Co   C    H
     5     1     4
Direct
  0.5110725781198682  0.4300455811962109  0.5336790702982914
  0.4288331557395777  0.4425488341188117  0.4494631228195974
  0.5356799907164490  0.4937313723296479  0.4347248409909834
  0.4145965117205000  0.5017957740572626  0.5523437150571772
  0.4396491504939399  0.5659694870750596  0.4533642507525203
  0.4980959167433147  0.5249545816035035  0.5148808651982163
  0.4480230754881377  0.4304742183120737  0.5918729370404100
  0.5717216618952679  0.4227494572544637  0.4736092107560319
  0.4984005102820168  0.5603012564987977  0.3910273065623750
  0.3766874488009362  0.5695194375541689  0.5112346805244027
```

```
Co6CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    19.3980482999999992    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.3980482999999992    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9003487999999997
   Co   C    H
     6     1     4
Direct
  0.4319483581439230  0.5077010544274898  0.6088210362562719
  0.5905025511780921  0.4755798916787150  0.5639235483647353
  0.4933895941193970  0.4158173378873967  0.5587170402100406
  0.4969986836449177  0.5225483226913388  0.5054366128634015
  0.5247280644341880  0.4653379570412077  0.6685971443675987
  0.5327886369898978  0.5695724616183502  0.6143823517812110
  0.4810953666366887  0.5550817641880638  0.4043285781557769
  0.5295235932150337  0.5484036209601338  0.3729723722503915
  0.4398176651796223  0.5234165433598403  0.3796571811936140
  0.4658577575417226  0.6097234893636452  0.4056528872812930
  0.5741097289165191  0.3949375567838159  0.5337612472756514


Co7CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    18.4042816999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.4957397000000014    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.2152725000000011
   Co   C    H
     7     1     4
Direct
  0.5135923649354314  0.4438005322056855  0.5758921222762662
  0.4884679073650813  0.5829086653022884  0.4685969117181086
  0.5797631212422045  0.5078521301223478  0.5042334260518611
  0.4479599307545575  0.3851304728950951  0.4964852172401364
  0.5030707289495006  0.5616705256216049  0.5801188899058441
  0.4195998701809442  0.4967944693185669  0.5193892258618579
  0.4952755871567857  0.4712353454383598  0.4312591628160254
  0.5483139144633520  0.4099993133586255  0.4908051123614722
  0.4739674978167169  0.5461871572428086  0.3955560833135288
  0.5058238165027960  0.5005481216603946  0.6374806016027773
  0.5878194246417970  0.3690475470722318  0.4793208442633946
  0.4928958359908341  0.6319157197619952  0.5352424025887246


Co8CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    20.1837008000000004    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.7661842000000014    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.3008431999999992
   Co   C    H
     8     1     4
Direct
  0.5166626135373938  0.5495364808519450  0.3961412631679205
```

```
  0.5384416331622691  0.5292660881792206  0.5935888972515012
  0.5791449985848346  0.4995352212462518  0.4833680928394674
  0.4418092023814037  0.4762938423829142  0.5655088432031200
  0.4446138867932594  0.5997345334463516  0.5854737107690509
  0.5255417932754880  0.6084823468898848  0.5031319651227029
  0.4832173754655048  0.4469678224216402  0.4590533319867448
  0.4291041685471906  0.5557726811415290  0.4726699187552106
  0.5295542631050199  0.4371619050587361  0.5487826658462854
  0.3824942230485471  0.5433368754805784  0.5537319410652222
  0.5474815393671373  0.3851489629582726  0.5673642195681635
  0.5958962939725648  0.5272430016566615  0.4009888032598999
  0.4397680087593873  0.5768402382860148  0.3883863471647148


Co9CH4 shown in Figure S5
1:P1(1)
   1.00000000000000
    19.4254281000000013    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.2027231000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9100204000000005
   Co   C    H
     9     1     4
Direct
  0.5821376730609057  0.4621799400972503  0.5152783919334365
  0.6227677849183063  0.5610926760081943  0.4583276631218274
  0.4636294488059008  0.4511567412137427  0.5490945065827304
  0.5327721352129525  0.5497029795281350  0.3747692015974068
  0.4435237061845814  0.4866753274074347  0.4249585974208192
  0.5033723276042011  0.4412624863151829  0.3259382950354094
  0.5112332157650658  0.5540474777375326  0.5016533831224657
  0.5119006539963595  0.3944173210668870  0.4447010335013400
  0.6037039288257556  0.4581822515065995  0.3910598683287883
  0.4487122186594975  0.5419300555638724  0.6002211084527199
  0.4663483948779436  0.5303387622324959  0.6576405417739954
  0.4687566962801514  0.5963832923498755  0.5905215461408203
  0.3921647956847937  0.5482601422490669  0.5995399228731382
  0.4684970201235624  0.3696405467237312  0.5219259401150931


Co10CH4(a) shown in Figure S5
1:P1(1)
   1.00000000000000
    20.0397916000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0844553999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.7691115999999987
   Co   C    H
    10     1     4
Direct
  0.4371852604892810  0.5165974089049405  0.4461705401595892
  0.5703550900507706  0.5164547908697492  0.5547225139732539
  0.4762220972013584  0.5824128268287320  0.5342644925702924
  0.5512515376956483  0.5628688587596056  0.4439230460633145
  0.4061317931609869  0.4348994520509117  0.5210860977030992
  0.3724673504717651  0.5392654645348549  0.5483350025294986
  0.4621016794942849  0.4953182991479603  0.6113736528676759
```

```
  0.4815584803588734  0.4043766650108184  0.4302635637255162
  0.5196010517507278  0.4184841304851869  0.5452006551756755
  0.5897638255352300  0.4494424967421967  0.4483495829413983
  0.5179422501557945  0.4850155616318222  0.4080360459048319
  0.5199582755098687  0.6318339909748975  0.4780808419538973
  0.4055238607069580  0.3805479974289483  0.4531804720328284
  0.5602882533082646  0.3727897739914683  0.4463751796053070
  0.6258491941101899  0.5206522826379085  0.4718683127938178


Co10CH4(b) shown in Figure S5
6:P1(1)
   1.00000000000000
    18.3906333999999987    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9742710999999993    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.8590495999999987
   Co   C    H
    10     1     4
Direct
  0.5380755864817491  0.5440707824610975  0.4633809765855155
  0.4516052538385032  0.5914563370821322  0.5371349777823531
  0.3400811068625765  0.5407020057477151  0.5514638861817354
  0.4222908978355536  0.4914304315171155  0.4691320368012051
  0.4752456730784905  0.3751773840754897  0.4851305702067279
  0.5834590909791196  0.4334064596369520  0.4994926359652003
  0.4333132325894078  0.5155504956678655  0.6283178763149256
  0.3906027220103725  0.4239467919037397  0.5612678159623233
  0.5100207125718353  0.4077714763206857  0.5930756961883822
  0.5473911229655690  0.5261912331178983  0.5807584056606632
  0.5136088265835731  0.4502271148167809  0.4234349332017326
  0.5156405403571492  0.5383594795427281  0.6590267583234776
  0.5154553837104188  0.4373686786491037  0.3690990559555791
  0.6040917675729011  0.4379011112217655  0.5822240645026667
  0.5456880825627869  0.6114902182389278  0.5245003103675098


Co1 shown in Figure S6
Co
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Co
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


Co2 shown in Figure S6
Co
   1.00000000000000
    22.4819999999999993    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
```

```
   Co
     2
Selective dynamics
Direct
  0.4564752980101576  0.5000027788023488  0.4999998465559107   T   T   T
  0.5435247019898425  0.4999972211976512  0.5000001534440894   T   T   T


Co3 shown in Figure S6
1:P1(1)
   1.00000000000000
    15.3978851999999993    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.1172958999999985    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.7433622000000000
   Co
     3
Direct
  0.4811453581448538  0.4457319525522470  0.5717563453442739
  0.5209831161069357  0.5571066827628768  0.5242685634665024
  0.4996315257482078  0.4696813646848720  0.4398950911892210


Co4 shown in Figure S6
1:P1(1)
   1.00000000000000
    16.4244383999999997    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.8488767999999993    0.0000000000000000
     0.0000000000000000    0.0000000000000000   15.7790736000000003
   Co
     4
Direct
  0.4285831604710672  0.4998434784543619  0.5628870572900799
  0.4616265759184647  0.4234811756452374  0.4646519042889078
  0.4616929870268221  0.5766351938164470  0.4650449571693674
  0.5613672765836504  0.5000501520839425  0.4580360812516456


Co5 shown in Figure S6
1:P1(1)
   1.00000000000000
    16.4671623999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.3406742999999999    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9877724999999984
   Co
     5
Direct
  0.5118542114074476  0.5823904445660973  0.5000075853716376
  0.4177568525648049  0.4504868506136837  0.4999893673391178
  0.5779572905782996  0.4347093667997594  0.5000028827944840
  0.5023459929175020  0.4891595348937926  0.5866403621425071
  0.5023656525319433  0.4891738031266609  0.4133598023522607


Co6 shown in Figure S6
2:P1(1)
```

```
   1.00000000000000
    17.9777777999999984    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9777777999999984    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9777777999999984
   Co
     6
Direct
  0.4107348085540009  0.5000000334571788  0.4999999755024617
  0.5892651752466673  0.4999999480484187  0.5000000336414565
  0.5000000455648342  0.4107347782117591  0.5000000015208413
  0.4999999564174281  0.5892652588188975  0.5000000103822367
  0.4999999983463513  0.4999999974359952  0.4107348186034885
  0.5000000158707180  0.4999999840277508  0.5892651603495153


Co7 shown in Figure S6
1:P1(1)
   1.00000000000000
    17.2434577000000004    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.2434577000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.4969086000000011
   Co
     7
Direct
  0.4689476614120146  0.5243390367069867  0.4482734217477323
  0.4842908040464295  0.5620296726196666  0.5726473899546932
  0.5099131625385728  0.4362922507946011  0.6069904910788799
  0.3990005085019960  0.4651576469939853  0.5411604896568349
  0.5828869761920756  0.4947328042096698  0.5157132008062186
  0.4974494680575875  0.3992913258741277  0.4817251562945081
  0.5575114192513309  0.6181672628009660  0.4761998504611266


Co8 shown in Figure S6
1:P1(1)
   1.00000000000000
    18.7411722999999988    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.6570110000000007    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.8412892999999997
   Co
     8
Direct
  0.5410715501408596  0.4773174389956786  0.5533240017787229
  0.4617004587594202  0.5780732064242913  0.5645371618732757
  0.4230794170479760  0.4615985299407522  0.5295568014495203
  0.5053530260246725  0.3962751445873814  0.4662941822052570
  0.6098578850634699  0.4605797376139481  0.4518133849381657
  0.5464354913009863  0.5712712905646724  0.4744936539495979
  0.4283219425906548  0.5561536159365635  0.4501147231066505
  0.5109602290719634  0.4910410359367230  0.3871060906988031


Co9 shown in Figure S6
1:P1(1)
   1.00000000000000
```

```
    18.3688108999999997    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.3688108999999997    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.4371146000000010
   Co
     9
Direct
  0.4791021779475105  0.4688724745588335  0.6126083629028793
  0.4977956394192584  0.5864493085769309  0.5745140670958285
  0.5340089016103639  0.3849259160579870  0.5310878311794890
  0.3802467525478460  0.5392747840281250  0.5741561534601048
  0.5840212388431519  0.5000338437062334  0.5502697653304274
  0.5310535578182600  0.4752332322644144  0.4376914876965479
  0.5673023998238721  0.5945208274754821  0.4630709658521390
  0.4259732650823609  0.4444423351791240  0.5000848739575193
  0.4449960669073740  0.5617372781528749  0.4618164925250725


Co10 shown in Figure S6
1:P1(1)
   1.00000000000000
    18.4334954000000018    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9993167999999990    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.0025991999999988
   Co
    10
Direct
  0.5000281043332541  0.4350047121031111  0.5000662275316154
  0.5000303271896751  0.5650359178791097  0.5000174114106689
  0.4886513377915139  0.5001372620794929  0.6105781021495768
  0.5113348113001901  0.4999165019168961  0.3894344581702137
  0.6004933304220049  0.4999031554429648  0.5432703744334693
  0.3995715298451841  0.5000775278955700  0.4567064347692442
  0.6105291045514832  0.4374505155459772  0.4332375853127338
  0.3894304431276012  0.4377073012570796  0.5667242078198305
  0.6105505460468850  0.5622771704891539  0.4332054232020980
  0.3893804653922083  0.5624899353906443  0.5667497752005464


Ni1CH4 shown in Figure S7
2:P1(1)
   1.00000000000000
    17.2535680999999990    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.1199234999999987    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.5883101000000011
   Ni   C    H
     1     1     4
Direct
  0.5075150488273362  0.4291621603692292  0.5470718878209934
  0.4821188825490715  0.5008173316961178  0.4741510119365976
  0.4435569818059948  0.4647073965331619  0.4373895919273399
  0.5533848109838364  0.4821764664191844  0.5908673384366395
  0.4498495552722242  0.5505200022047957  0.4981456620686403
  0.5333647205615403  0.5206266427775146  0.4421945078097982
```

Ni2CH4(a) shown in Figure S7
1:P1(1)
   1.00000000000000
    17.7577795000000016   0.0000000000000000   0.0000000000000000
     0.0000000000000000   18.1459131000000014   0.0000000000000000
     0.0000000000000000   0.0000000000000000   18.8912821000000015
   Ni   C   H
     2   1   4
Direct
  0.4469271657579190 0.4371669122767768 0.6011305821812418
  0.4883504366173640 0.4455368219165818 0.4942583744504087
  0.5280560190072506 0.5245532886569331 0.4398334926109813
  0.5874423645676503 0.5125408553854076 0.4262288057209495
  0.4949842472074901 0.5293639863995412 0.3903797993015506
  0.4483581090909398 0.3751027762969006 0.5466634576642340
  0.5256316577513810 0.5775153590678688 0.4682854880706327

Ni2CH4(b) shown in Figure S7
28:P1(1)
   1.00000000000000
    17.6033684000000008   0.0000000000000000   0.0000000000000000
     0.0000000000000000   18.3560201999999997   0.0000000000000000
     0.0000000000000000   0.0000000000000000   17.5942563999999990
   Ni   C   H
     2   1   4
Direct
  0.4510603480523001 0.5241240551106026 0.4840859026235030
  0.5596668150319716 0.4906566917058415 0.4199307919078740
  0.5274399338337002 0.4668322956524471 0.5153938165339590
  0.5633003787899975 0.4887500349952077 0.5616012182957604
  0.5136845097821682 0.5624771292334966 0.4289605141528652
  0.4008662152129489 0.5417586045534711 0.5498895170553465
  0.5095617992969045 0.4090711887489299 0.5205382394306951

Ni3CH4(a) shown in Figure S7
1:P1(1)
   1.00000000000000
    17.8243133000000000   0.0000000000000000   0.0000000000000000
     0.0000000000000000   17.8966725999999987   0.0000000000000000
     0.0000000000000000   0.0000000000000000   17.6065284999999996
   Ni   C   H
     3   1   4
Direct
  0.4291058085854439 0.5242994041991638 0.5398866386183958
  0.5128877081698658 0.4305411889255988 0.5218538472778367
  0.6117904549993898 0.4876768931172625 0.4657258087616062
  0.5154321998913793 0.5273009015120470 0.4795159809408634
  0.4941293571160438 0.5379546885762453 0.4209819154864269
  0.5383747374538287 0.5776533903627006 0.5112038510400856
  0.4264598337773595 0.4356482772370701 0.5494808319931775
  0.5890299000066895 0.4027852560699163 0.4804911258816109

Ni3CH4(b) shown in Figure S7
2:P1(1)
   1.00000000000000
    17.6886958000000014   0.0000000000000000   0.0000000000000000
     0.0000000000000000   17.3374531000000012   0.0000000000000000
     0.0000000000000000   0.0000000000000000   18.2271503000000017
   Ni   C   H
     3   1   4
Direct
  0.4332220492965401 0.5065308126424763 0.5391362027029334
  0.5073483730444602 0.4177606573077243 0.4672716833475012
  0.5522844119671604 0.5471555579621087 0.4882593348622988
  0.5269325116942324 0.4709012069136867 0.5485391797797502
  0.5563181100980514 0.4509181034573171 0.5985620387439604
  0.5625772315082104 0.4798242544520397 0.4251401099321545
  0.4174709916347595 0.4310900789865383 0.4864208276135664
  0.4723063207565811 0.5880293282781147 0.5130606230178404

Ni3CH4(c) shown in Figure S7
34:P1(1)
   1.00000000000000
    17.0670855999999986   0.0000000000000000   0.0000000000000000
     0.0000000000000000   17.8202549999999995   0.0000000000000000
     0.0000000000000000   0.0000000000000000   18.7781641000000015
   Ni   C   H
     3   1   4
Direct
  0.5172385739676189 0.4638515495475276 0.4350999634145741
  0.4650879875875852 0.4597705976343339 0.5438520000459885
  0.4194078329636940 0.3837107806019647 0.4557979371428734
  0.5222940847179715 0.5470810308706680 0.5047834540698545
  0.5690774082543549 0.5463551218232234 0.4601647616931770
  0.4845043873444467 0.5959563839888312 0.4943800338977097
  0.4937940163424317 0.3834779733413016 0.4016104624803030
  0.5592057088218938 0.5580565621921419 0.5521213872555156

Ni3CH4(d) shown in Figure S7
41:P1(1)
   1.00000000000000
    17.7498481999999989   0.0000000000000000   0.0000000000000000
     0.0000000000000000   17.6876728999999990   0.0000000000000000
     0.0000000000000000   0.0000000000000000   18.2236876000000017
   Ni   C   H
     3   1   4
Direct
  0.5064840931864560 0.5044108805391397 0.4090204507552674
  0.5514516475842116 0.4523486074609402 0.5166646135996698
  0.4282871482686136 0.5910146971623434 0.4619375017063173
  0.4942015618720534 0.5323528154906996 0.5008841580622412
  0.5428515760493026 0.4018842132214375 0.5901784595785894
  0.5734379635680591 0.4367132902714501 0.6012845777704404
  0.5646178895026006 0.4382242206387106 0.4265652952237406
  0.4355981199687040 0.5582812752152868 0.3782949433037350

```
Ni4CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    18.1683641000000016    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.3233855000000005    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1683641000000016
   Ni   C    H
     4     1     4
Direct
  0.4326904893675270  0.5000498408017875  0.5673310727416384
  0.5673312426308004  0.4998984167403372  0.5673012708029560
  0.5672939428782483  0.5000141911158469  0.4326753288790415
  0.4326631473251486  0.4999005975514295  0.4326909004389410
  0.5000162983228649  0.4995644824980270  0.5000307656706229
  0.5000172997104306  0.5001572430156361  0.6286435808731790
  0.4999782558882815  0.5001195315465558  0.3713454622096613
  0.6286446173073541  0.5001461331646303  0.4999819527336979
  0.3713547065693412  0.5001495635657418  0.4999996656502617


Ni5CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    19.3684511999999991    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.3684511999999991    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.8980412000000015
   Ni   C    H
     5     1     4
Direct
  0.5363339227248672  0.4949001450284539  0.5885789691461821
  0.4400200924287058  0.4503551419727090  0.4445648663892055
  0.5597909018792158  0.4388323748219163  0.4721784820477997
  0.4497176581767309  0.5724890208434382  0.4537008546243227
  0.5694354506887450  0.5609259410594940  0.4808949746300918
  0.4943362968096776  0.5041425073567585  0.5000816345334637
  0.6237252435247700  0.4950261574436428  0.4782192309721705
  0.3892879777862969  0.5170670984172174  0.4294501575606698
  0.5154604348634670  0.6253767767050211  0.4641774170693131
  0.4964320211175154  0.3867448363513598  0.4460434130267846


Ni6CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    19.7639341000000002    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.0147875999999982    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.4859945000000003
   Ni   C    H
     6     1     4
Direct
  0.4799410738125303  0.4383020465314951  0.5003968021221215
  0.4144622656136084  0.5369247059849858  0.5507033809436707
  0.5112951963742292  0.5893915549449508  0.5914977182796195
```

```
  0.5570093228846384  0.5939858209995805  0.4673275380190951
  0.4565220623965344  0.5376793722993450  0.4249268078256006
  0.5808879483566913  0.4942845928990587  0.5422110068595345
  0.5001699009680024  0.5316628184978612  0.5119177303769953
  0.5017248617299014  0.6069159879151195  0.4017111806832449
  0.4235414318365843  0.4584836590319751  0.4373370008228835
  0.5473941694368423  0.4148984942944993  0.5469141168857856
  0.6202217665904387  0.5680509466011243  0.5198267171814439


Ni7CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    18.7853664999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.7422816999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.4798617999999983
   Ni   C    H
     7     1     4
Direct
  0.5270570458193726  0.4877410973046966  0.4202006532099718
  0.5988136977789420  0.4950290866951603  0.5274597101015718
  0.5245765276759286  0.5800203217617539  0.5760052932333412
  0.4067248166260135  0.6155590640980531  0.5742364250660051
  0.4271475805601212  0.4997107501089936  0.5758059855933746
  0.5018452714792879  0.4162807807463662  0.5224775998515775
  0.4547179705789018  0.5766569721316216  0.4662409112310224
  0.5021310237203451  0.5087942216815852  0.5134875089903210
  0.4810868001147909  0.5487775139453053  0.3852117645197964
  0.4343682821398500  0.4153244610319930  0.5768745936109205
  0.5776249091136729  0.4263038609598342  0.4671174855464085
  0.6106960743927746  0.5584418695346409  0.5810720690456912


Ni8CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    19.9566562999999988    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0737057000000014    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.8081777000000017
   Ni   C    H
     8     1     4
Direct
  0.4820747204366997  0.5664034821068891  0.4423642250953875
  0.4810329433134218  0.4700508631128076  0.3724592963552072
  0.5726388366223447  0.4895248007357360  0.4472869349147346
  0.4069871078624415  0.4774830748824138  0.4664741286273316
  0.5640120427834094  0.4659063081040847  0.5640691963965151
  0.4971559133888371  0.5674860556269486  0.5594494569581594
  0.4455483934554305  0.4585956831567750  0.5780746586616676
  0.4974646820886030  0.4049259534912350  0.4723406806774819
  0.4945910484228288  0.4943877722708814  0.5034414111589773
  0.4867068075180667  0.6233868300023822  0.5020059812263240
  0.3740372860744633  0.4541720460704926  0.5380799317355612
  0.6240304054828852  0.4707253153881711  0.5084592337513570
  0.5114298125505637  0.4587218150511751  0.6270448644412810
```

```
Ni9CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    18.3309004999999985    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.8813788000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.8351550000000003
   Ni   C    H
     9     1     4
Direct
  0.5666411598800297  0.5836870854340525  0.4849787408238317
  0.3956572165671721  0.5597733641740957  0.5173959709648449
  0.4458486047636315  0.4515178211455850  0.5686345066881833
  0.5470130168146345  0.4730262509710738  0.4206157479184849
  0.4983417708361445  0.5690723120574398  0.5850620149175461
  0.4641375320674150  0.5742622749705343  0.4172390635063027
  0.4206972130501797  0.4552183035692182  0.4445216287292995
  0.4689408780269024  0.6618531916674524  0.5063425875696720
  0.5723312883799235  0.4691547253624752  0.5447716431011034
  0.5011284778431652  0.4282445296492003  0.4927063899428062
  0.5179824872243591  0.4823354996283313  0.6187607837388026
  0.4681321866600318  0.4901092360261475  0.3734068485865535
  0.6181240200069684  0.5038810747835056  0.4725879564987006
  0.3681941478794197  0.4687143305608754  0.5196161170138530


Ni10CH4 shown in Figure S7
1:P1(1)
   1.00000000000000
    19.5831474000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.9440087000000013    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.6400028999999989
   Ni   C    H
    10     1     4
Direct
  0.4775333931098745  0.4723653453487488  0.6158024543531251
  0.4961460042155722  0.4901331086543789  0.3864660154854164
  0.5349260054732651  0.6071452921542733  0.4255186691007878
  0.4749914129760356  0.6595331504213460  0.5140084405142654
  0.5650164139213031  0.4283419435722197  0.5421708366592148
  0.4480198561045511  0.4608659148794243  0.4972094780481303
  0.6042325205350816  0.5038402168023707  0.4432939539317534
  0.4147451929576571  0.5643868014303239  0.5663868868668307
  0.5275216045047263  0.5511067984578014  0.5325282692534872
  0.4238307285726809  0.5735292558649138  0.4473832048192755
  0.5325205525182729  0.4488932798060861  0.4605454573450694
  0.4152973559852352  0.4882004638350387  0.4160939672653333
  0.5359277706909000  0.4128226498572080  0.6191224645734995
  0.4096611381840852  0.5170965189824300  0.6350809742624709
  0.5694700502507581  0.5450992599334430  0.3721989275213332


Ni1 shown in Figure S8
Ni
```

```
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Ni
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


Ni2 shown in Figure S8
Ni
   1.00000000000000
    22.3279999999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Ni
     2
Selective dynamics
Direct
  0.4534011308175732  0.4999999894465211  0.4999999378694742   T   T   T
  0.5465988691824268  0.5000000105534789  0.5000000621305258   T   T   T


Ni3 shown in Figure S8
1:P1(1)
   1.00000000000000
    16.5430497000000010    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.5430497000000010    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.5430497000000010
   Ni
     3
Direct
  0.4526933539804298  0.4530265950043398  0.5469732816418664
  0.5469734051847674  0.5473068027511295  0.5469733681007346
  0.5469732408348064  0.4530266022445272  0.4526933502574026


Ni4 shown in Figure S8
1:P1(1)
   1.00000000000000
    16.2082720000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.3951728999999986    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.3542075999999987
   Ni
     4
Direct
  0.4657452268003004  0.5003620988839091  0.5623485826704370
  0.4313949064495351  0.4995502433452537  0.4320896990196043
  0.5515798549496197  0.4329421946911687  0.4708962944706930
  0.5512800118005448  0.5671454630796686  0.4699654238392637


Ni5 shown in Figure S8
```

```
1:P1(1)
   1.00000000000000
    16.5423073000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.0846145000000007    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.1052423000000005
   Ni
     5
Direct
  0.4606352447987898  0.5009247785066492  0.4188387103114701
  0.4440884314974975  0.4990012370581020  0.6066553334740771
  0.4533701145509173  0.4104813452983893  0.5118250220833725
  0.4529561808746336  0.5894645983177083  0.5136353443495124
  0.5490900282781629  0.5001280408191513  0.5208855897815695


Ni6 shown in Figure S8
2:P1(1)
   1.00000000000000
    17.4706423000000015    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9068047999999997    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.8972994999999990
   Ni
     6
Direct
  0.5000199581679818  0.4999576960359549  0.4027026374868679
  0.4999977813499439  0.5000454988986927  0.5973002968011475
  0.5226768701891187  0.4112497684410685  0.5000122415289942
  0.4773035151393075  0.5887469928676650  0.4999823590802775
  0.5908953181414659  0.5221911089078223  0.5000013451391666
  0.4090965570121790  0.4778089348487968  0.5000011199635533


Ni7 shown in Figure S8
1:P1(1)
   1.00000000000000
    18.9501516999999993    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.3690791000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.5222487999999998
   Ni
     7
Direct
  0.4797429933399595  0.5166620789901667  0.3992143711286942
  0.4931612436621299  0.4592208958195972  0.5684772892740280
  0.6125373250302003  0.4836254490045732  0.5658191998360544
  0.4368885277874558  0.4126959580334315  0.4646031209798904
  0.4154398241687410  0.5374587601001497  0.5036812880828601
  0.5576743619921189  0.4379427587202140  0.4635739091839335
  0.5366057240193857  0.5624240993318739  0.5024708215145349


Ni8 shown in Figure S8
1:P1(1)
   1.00000000000000
    18.7710052000000012    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4770582999999995    0.0000000000000000
```

```
     0.0000000000000000    0.0000000000000000   17.9749133000000008
   Ni
     8
Direct
  0.4999400660088171  0.5780174440024567  0.5887589274809152
  0.4114878438115784  0.4917633454553074  0.4160548577696470
  0.5323047919928435  0.4553589888772319  0.6006088055962048
  0.4324581677223602  0.5983684080842200  0.4840935793063137
  0.5203322506059416  0.5551119013891611  0.4118400240342076
  0.5126750709719290  0.4431670919220286  0.4731306755892895
  0.5880175520122777  0.5351447606508094  0.5165127861883828
  0.4240042568742572  0.4866380596187813  0.5454203440350459


Ni9 shown in Figure S8
1:P1(1)
   1.00000000000000
    19.1931723000000005    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4268600999999990    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.5005245000000009
   Ni
     9
Direct
  0.5931621673703871  0.5765441101974037  0.5646043369183062
  0.5509662792611868  0.5643671687894282  0.4452723071091211
  0.4624272255692224  0.4262603492317193  0.5042881846805507
  0.3874894925283536  0.5110845596115090  0.5591228992218137
  0.5217173013070965  0.4506690794128871  0.3952660014557635
  0.4344188033590067  0.5273197236812119  0.4426500949608932
  0.5029760905965485  0.4977560984162723  0.6004680977283237
  0.4749810241303653  0.5986831373030128  0.5386752867108988
  0.5789616158778347  0.4635057733565590  0.5072427912143265


Ni10 shown in Figure S8
1:P1(1)
   1.00000000000000
    19.4415624999999999    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.4606050000000010    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.4562092000000000
   Ni
    10
Direct
  0.4998351206772356  0.5004564111472346  0.5922770583285608
  0.4998456110532469  0.4995126956728356  0.4077075633791840
  0.5002678202417528  0.4028052509434604  0.5004434781341470
  0.4999969523033758  0.5972307096610512  0.4996668872663240
  0.4174384307091956  0.5911001767458244  0.4130695778041973
  0.5825654232427175  0.4084812320900408  0.4135407183819218
  0.5824677051316224  0.5916533259880011  0.5864662517412146
  0.4175983504359981  0.4088025519338083  0.5869867148795466
  0.4125041194957291  0.4998042031451638  0.4999413311073734
  0.5874804667091191  0.5001634426725758  0.4998904189775275
```

Cu1CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    17.1831518000000010   0.0000000000000000   0.0000000000000000
     0.0000000000000000   16.8141315999999996   0.0000000000000000
     0.0000000000000000   0.0000000000000000   17.8101078999999984
   Cu   C   H
     1   1   4
Direct
  0.5181271106657912 0.5669135208910140 0.6177117303184755
  0.5064244194525797 0.4733580848170377 0.4665753397951291
  0.5411831588581015 0.4452077849324846 0.5112532601155436
  0.5176582661909978 0.4423454253899262 0.4132963804285089
  0.5232835111420249 0.5361280026326202 0.4609328692587743
  0.4443835336904974 0.4691371813369171 0.4808204200835670

Cu2CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    18.7505567000000006   0.0000000000000000   0.0000000000000000
     0.0000000000000000   17.1841557000000016   0.0000000000000000
     0.0000000000000000   0.0000000000000000   20.1794468999999985
   Cu   C   H
     2   1   4
Direct
  0.4692315747172713 0.5150724055060872 0.5133611925692471
  0.4134337747671795 0.5423619573377865 0.6082426365422879
  0.5262535796738690 0.4859511775482543 0.4163952613719615
  0.5828460236101605 0.4973104271370260 0.4068337056933143
  0.5002381016695873 0.5433730171221092 0.4272006748199012
  0.4996370175764306 0.4609873137886478 0.3729643140084135
  0.5228299279855049 0.4426237015600935 0.4575722149948687

Cu3CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    17.8225633999999999   0.0000000000000000   0.0000000000000000
     0.0000000000000000   18.6995880999999997   0.0000000000000000
     0.0000000000000000   0.0000000000000000   18.2994306000000009
   Cu   C   H
     3   1   4
Direct
  0.5860338026270770 0.4628908460631438 0.4319519335096825
  0.4883853077686538 0.4696947207037577 0.5159333699261175
  0.5477299473971616 0.5749776808179581 0.4817035095352994
  0.4587981685016502 0.5647742507817547 0.5587516423046887
  0.5271906083843231 0.4039569635923143 0.4729833940279006
  0.3982205146681980 0.5654335989439755 0.5465196149899313
  0.4756624974189712 0.6220087175568690 0.5504987475586618
  0.4680691532339679 0.5556032215402293 0.6175777881477158

Cu4CH4 shown in Figure S9

```
1:P1(1)
   1.00000000000000
    18.3974483999999983    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.0655704999999998    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.0974781000000000
   Cu   C    H
     4     1     4
Direct
  0.5402144442059982  0.4959292674685064  0.6262597396602062
  0.5344307620220339  0.4895889451997023  0.4167645174221759
  0.4713806219546999  0.4912125694253280  0.5232339896552292
  0.6027124463909144  0.4906328865730161  0.5194510823016414
  0.4253597329468591  0.4895243515989877  0.4232531006766254
  0.6216395595066502  0.4901983568147368  0.4342801976067424
  0.4131342441426393  0.5361034103843532  0.3877554564105341
  0.3850255067996848  0.4992115858453154  0.4669275336546548
  0.4072326820305222  0.4371786266900578  0.3994343826121867


Cu5CH4(a) shown in Figure S9
1:P1(1)
   1.00000000000000
    18.4062005000000006    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.3416518000000011    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.9014707999999985
   Cu   C    H
     5     1     4
Direct
  0.5083947482479918  0.4330222785869378  0.4896343302963131
  0.6000880232079961  0.4547551451122961  0.5789488916474287
  0.5125620313292995  0.6546306133343636  0.5373312510408391
  0.5809332452386605  0.5693570718722295  0.5876735557013750
  0.4869764927093094  0.5487794163345914  0.4948952396114144
  0.4373641494250622  0.4828599823647192  0.4369242534490844
  0.3797072353864923  0.4755382579824096  0.4511813872210995
  0.6334527642714636  0.5145029744491192  0.6258261902835257
  0.4439040255896410  0.4883283215392755  0.3759707154634668
  0.5701472845940799  0.3910759384240441  0.5347541852854402


Cu5CH4(b) shown in Figure S9
3:P1(1)
   1.00000000000000
    17.5010477000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.7149365000000003    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.3234825000000008
   Cu   C    H
     5     1     4
Direct
  0.4483770281929231  0.5353361602457823  0.6141643576998916
  0.5024614773506706  0.4789583901810194  0.5200498020335396
  0.5399900939567445  0.5458957171084571  0.4297777040599972
  0.4838955359128770  0.6039621589024020  0.5204687506374277
  0.4006935281612676  0.6415694582537744  0.6063534455431626
  0.5463272353749606  0.4500232539556384  0.4223390686915653
```

```
  0.5267707249868269  0.4527037591508223  0.3678154804225431
  0.5112256721809975  0.4093764063849178  0.4431215828055952
  0.5282554678464377  0.6212978236356675  0.4462173417388819
  0.6062532360362970  0.4340568721815135  0.4237324663674050


Cu6CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    17.6545668999999990    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.4939190000000018    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1214474999999986
   Cu   C    H
     6     1     4
Direct
  0.4657093069614404  0.4769384298543420  0.3514359627785454
  0.5834090784701902  0.4254847953571044  0.3228894898811003
  0.5638861896689586  0.4659048640321833  0.4435597464353733
  0.4008593229658022  0.5734828800459228  0.4219673127697767
  0.3385978485453993  0.5056726161165541  0.3299704203724065
  0.4962418756319245  0.5584817462733057  0.5107426207199438
  0.5708106460317004  0.4843776010064818  0.5543259211165257
  0.5411201784594952  0.5137579215291609  0.6013195819207133
  0.6311240189215306  0.4989943886123984  0.5614056745833792
  0.4353357996770242  0.6228089022861569  0.4907623203313775
  0.5620357346665308  0.4232058548863880  0.5671209490908471


Cu7CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    18.3937282999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4469385000000017    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.9267842000000002
   Cu   C    H
     7     1     4
Direct
  0.4958768487119755  0.4706141059415270  0.6055291346503302
  0.4068749087396846  0.5394054320224601  0.5347977787286765
  0.6109057883038765  0.5369565745491628  0.6183469671496881
  0.5831639319121703  0.4984098319568266  0.5093231873290290
  0.5168502027534917  0.5968716507896153  0.5619814139020263
  0.3819964287568655  0.4187236131643675  0.5631122097167535
  0.4737148158499195  0.4393966277385003  0.4835318963529550
  0.5633406163424649  0.4400654583150937  0.4238129566598740
  0.6103063475311841  0.4767880019731341  0.4126338913563446
  0.5843363618498701  0.3842552956406117  0.4223738024746725
  0.5269842801719616  0.4496599150294990  0.3804830963268465
  0.4346494690765364  0.6227134928792032  0.5430536653528099


Cu8CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    18.0519405000000006    0.0000000000000000    0.0000000000000000
```

```
     0.0000000000000000   19.0026563999999993    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.4165841000000015
   Cu   C    H
     8     1     4
Direct
  0.4442757594875963  0.6085670840397154  0.5482405290126762
  0.4542658619073837  0.5406328068715509  0.4540066998757932
  0.5352523724095083  0.4239524843347282  0.6270553332362893
  0.6346841347831877  0.4903980710909773  0.5781073035435550
  0.5290812444734557  0.5545860716178953  0.6226512737443851
  0.4240044722013037  0.4891240036259286  0.5941730855753455
  0.5259813788601613  0.4602923413194399  0.5160869344135844
  0.5743600380268836  0.5838436078236131  0.5174048283561242
  0.4916143293755077  0.4466345116253204  0.4214389890908006
  0.4697068501281627  0.4800967630527568  0.3799689942863576
  0.5469016276436329  0.4310137942680995  0.4039175528504967
  0.4130476971994034  0.6136944506597372  0.4693080847625856
  0.4547042335038149  0.4006840096702486  0.4240603912520023


Cu9CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    21.1116194999999998    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.4229258000000016    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.1712597000000002
   Cu   C    H
     9     1     4
Direct
  0.5292445555767901  0.5157615550929948  0.6195812046804432
  0.5745333540064047  0.5345851737498477  0.3968275342098894
  0.5754498546578636  0.6273841664812531  0.5843809070554146
  0.5834123606629007  0.5266412786143591  0.5155268804584598
  0.4801154076318310  0.4644332822841716  0.5213384202278338
  0.5167794485622176  0.4321119355724194  0.4140582764830763
  0.4720042789524785  0.5877487450587966  0.5460235724543469
  0.5508730720163530  0.6332608195748303  0.4669092989332349
  0.4652911165502973  0.5533568923704618  0.4346573972424388
  0.4675920807389526  0.3689751070365999  0.4729987780181204
  0.4421087354808102  0.3735074741851155  0.5217587285616659
  0.5586296961315267  0.4641042015246039  0.3540242902989377
  0.4998437465621428  0.3249686210552059  0.4798328547512108
  0.4293422924694269  0.3549807473993623  0.4381618566249225


Cu10CH4 shown in Figure S9
1:P1(1)
   1.00000000000000
    21.0435583000000008    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.6145478000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.8599962999999988
   Cu   C    H
    10     1     4
Direct
  0.5336324531354647  0.6306335231423521  0.5190711289185886
```

```
  0.4332011482599501  0.6384097701116551  0.4537487544936070
  0.4691675630708542  0.5357674048256227  0.4981711962564032
  0.4915371603097102  0.4234323173516909  0.4963365391674175
  0.6047006089049557  0.4188926819335075  0.4685239843569760
  0.5937774552548238  0.5304738040363145  0.4915798921299322
  0.5313291145026964  0.3554046911842519  0.4094033553768242
  0.5262104514346061  0.4733127263105310  0.3919757733143635
  0.5313500012561985  0.5881468696424254  0.4087550179530935
  0.4310631320819936  0.5434598501150147  0.3817459300112237
  0.4354470460971864  0.4648146982579323  0.5649930890671382
  0.4124581386719391  0.5132156322227236  0.5730025041985549
  0.4606429949685907  0.4538390704287245  0.6121531159945556
  0.3949815134534503  0.4315125533915205  0.5574359218361447
  0.6468812185975827  0.4710044070457364  0.5078937969251685


Cu1 shown in Figure S10
Cu
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Cu
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


Cu2 shown in Figure S10
Cu
   1.00000000000000
    22.6039999999999992    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Cu
     2
Selective dynamics
Direct
  0.4509056249487535  0.5000000046534760  0.4999996252260545   T   T   T
  0.5490943750512465  0.4999999953465239  0.5000003747739457   T   T   T


Cu3 shown in Figure S10
1:P1(1)
   1.00000000000000
    16.9003157000000002    0.0000000000000000    0.0000000000000000
     0.0000000000000000   15.9875529000000007    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.1067965999999991
   Cu
     3
Direct
  0.4377240702973162  0.4585083285248526  0.4579825121410757
  0.5530912607502693  0.5326851091959025  0.4476977240945899
  0.4698046689524132  0.5331765622792496  0.5674097637643338
```

```
Cu4 shown in Figure S10
2:P1(1)
   1.00000000000000
    17.5102754000000012    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.1739522999999998    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.0188403000000008
   Cu
     4
Direct
  0.5696974094749020  0.5646595732614754  0.4124013927405887
  0.4519468977279982  0.5686966510205095  0.4844910290435813
  0.5181014958348334  0.4551477248302845  0.4834150393633061
  0.4001141969622694  0.4604760508877236  0.5560925388525300


Cu5 shown in Figure S10
1:P1(1)
   1.00000000000000
    17.1444019999999995    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.3988915999999989    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.1458164000000011
   Cu
     5
Direct
  0.4316222998539431  0.5532396180401982  0.3890565897006938
  0.5679831166963552  0.5531000477530700  0.6110607229810947
  0.5001225241928665  0.5535753444595567  0.4999134005623917
  0.5375895281088066  0.4274993147295727  0.5553832866721374
  0.4626925311480318  0.4275856750175961  0.4445760000836864


Cu6 shown in Figure S10
1:P1(1)
   1.00000000000000
    18.3426481999999993    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.6575105000000008    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.4879102999999994
   Cu
     6
Direct
  0.4186270522252573  0.5135752783511481  0.4939695172820323
  0.4520377988413984  0.6057487043848421  0.4046877478963088
  0.3894239871823014  0.4199090390399327  0.5831808649442862
  0.5443817038787221  0.5322346762957036  0.4592053622334278
  0.5122445242022940  0.4367548941893712  0.5518186380041032
  0.6333249336700207  0.4563774077389998  0.5165478696398441


Cu7 shown in Figure S10
1:P1(1)
   1.00000000000000
    19.0890768000000008    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.6187037999999987    0.0000000000000000
```

```
     0.0000000000000000    0.0000000000000000   16.2919741000000009
   Cu
     7
Direct
  0.4793579106935140  0.5103310657615691  0.4276860458717566
  0.5065254142462825  0.5281685603224738  0.5803933625003787
  0.4953672206277366  0.6285405099609828  0.4858986075163288
  0.3934939284155171  0.5583158116491499  0.5229385704169066
  0.4283502838111921  0.4339973182761079  0.5319179381755976
  0.5525923904684887  0.4275049081091429  0.5031217560303340
  0.5941528517372637  0.5476718259205645  0.4753837194887000


Cu8 shown in Figure S10
1:P1(1)
   1.00000000000000
    18.3771178000000006    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.7522376999999985    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.7832098999999992
   Cu
     8
Direct
  0.4538138314380972  0.5211914901878379  0.4216366170635814
  0.5492439468514013  0.4291825550532669  0.4500624725065810
  0.4247734208683795  0.3944974305403523  0.4622954589025585
  0.4954028365376312  0.5709540574527471  0.5430839571024711
  0.3828568650178108  0.5063787142016235  0.5352194655344548
  0.6033784271466978  0.4941359497705019  0.5559332057515168
  0.4891460765672238  0.4357888283968597  0.5729255963066250
  0.5758545955727411  0.5622409743968169  0.4361832268322170


Cu9 shown in Figure S10
1:P1(1)
   1.00000000000000
    17.5574830999999989    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.3483245000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.5006233999999985
   Cu
     9
Direct
  0.5128400478619231  0.5000809308541871  0.6159098988949711
  0.4940676829710904  0.3878500100750763  0.5387915950365704
  0.6053062031276695  0.4034173298475301  0.6176755033364576
  0.5915392209355974  0.4763767270535057  0.5032167170620184
  0.5662035844646498  0.5485190649345805  0.3933436785571503
  0.4281131339150289  0.5629393893203817  0.4047490568916338
  0.4112468147814463  0.4930234537689402  0.5209402783369415
  0.4863477155623914  0.4433464846407620  0.4161616580468299
  0.5107655963801924  0.5854466095050398  0.5138116138374195


Cu10 shown in Figure S10
1:P1(1)
   1.00000000000000
```

```
    18.7947130000000016    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.8341533000000005    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0018908999999994
   Cu
    10
Direct
  0.6014964179730339  0.4875840736977455  0.5704040672056636
  0.5569180787205110  0.5348626620573627  0.4645493755390906
  0.5244630305004748  0.4364083026002247  0.3891808520938493
  0.5136718043784284  0.4138915877437412  0.6316463211783380
  0.4810539178091037  0.5631736780971003  0.3647295172683480
  0.5250168638440216  0.4076593019757823  0.5073187890966687
  0.4293119224335275  0.4852057639856283  0.4560733591411725
  0.4551647701966812  0.6147925819461609  0.4751304699599106
  0.4099836861995818  0.4148052271070188  0.5578997870591138
  0.4797395079446398  0.5271568207892271  0.5648874614578295


Zn1CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    16.4673252999999988    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.6943711000000015    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.6126010000000015
   Zn   C    H
     1     1     4
Direct
  0.5301079576858307  0.4220461496554087  0.6408376137619801
  0.4674572963134472  0.5305262938736169  0.4848286527073884
  0.4474720884551068  0.5646339144274418  0.4349558736443970
  0.4490458593949384  0.5618209316195861  0.5368915137850744
  0.4396127949433157  0.4706980343249649  0.4841616951045494
  0.5338240032073600  0.5243746760989828  0.4842246509966101


Zn2CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    17.7773213000000005    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.4078541000000016    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.3625182000000002
   Zn   C    H
     2     1     4
Direct
  0.4268340700455489  0.5805250103949329  0.5793577159647983
  0.3808698605731023  0.5950802243752221  0.4090888893994807
  0.5556223191456311  0.4844462195603398  0.4616698342833001
  0.5531518645390900  0.5474731466660289  0.4624669512403011
  0.5794244259528322  0.4634188435847451  0.5131163244312242
  0.4982624277841064  0.4620424266161073  0.4553736539927580
  0.5903450319596908  0.4653941288026227  0.4155666306881339


Zn3CH4 shown in Figure S11
1:P1(1)
```

```
   1.00000000000000
    17.7415784999999993    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.3742282000000010    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.5211314000000016
   Zn   C    H
     3     1     4
Direct
  0.4136005294127360  0.5747142676588670  0.3791048297100110
  0.5000040158169347  0.4212783376536223  0.3787732708891524
  0.5864166914837052  0.5747246816298961  0.3791210814513577
  0.4999948304762922  0.5222272666244423  0.5707510842766260
  0.4999988085223355  0.5220363645395139  0.5077631794253789
  0.5505269016298627  0.5522447254795797  0.5911055713316786
  0.4494491713215025  0.5522576729960146  0.5911147463968219
  0.4999990513366351  0.4627166834180728  0.5916062365189726


Zn4CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    18.2831246000000007    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.8070534000000009    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.7859979999999993
   Zn   C    H
     4     1     4
Direct
  0.4996828962429912  0.4924400970955106  0.6134525587397162
  0.5154964726669992  0.6242153745755415  0.5133923959348887
  0.3736412348924363  0.5335816335661687  0.5334253812772265
  0.4115826830518000  0.3731914065092978  0.5354588365905022
  0.5401167741783676  0.4640614915762190  0.4310266468995789
  0.5384996372961229  0.4848341251327570  0.3764751516964794
  0.4899322833895401  0.4803357359066554  0.4590595617929794
  0.5430340395466646  0.4056687380834851  0.4299362700461742
  0.5879839787350828  0.4854913975543587  0.4590131970224514


Zn5CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    20.9517574999999994    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.3206592000000015    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.0915318999999997
   Zn   C    H
     5     1     4
Direct
  0.4487256491020556  0.4448608675661601  0.6107689796955522
  0.4709001685936044  0.4165802020413952  0.4530193630396352
  0.4434902085114348  0.5517478782711841  0.4891766635323280
  0.3486066518214071  0.4538653047090898  0.4877449460303653
  0.3992936859516559  0.5075533673756650  0.3445009301919703
  0.6023545309782752  0.5100832934736974  0.5547528195481543
  0.5896160283031731  0.4922284128565310  0.6074921629833996
  0.6516650815493421  0.5291720435448154  0.5541461556169840
  0.5697268682501270  0.5519302778180291  0.5392053897758459
```

```
  0.5971311269389256  0.4671083523434357  0.5174925895857622


Zn6CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    18.1930267000000008    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4175874999999998    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.8004911999999997
   Zn   C    H
     6     1     4
Direct
  0.5233060372070852  0.4861870007782687  0.6450368196389284
  0.4898218190327548  0.6232647480590843  0.5661750550647556
  0.4556199785707142  0.3670905268877747  0.5540758940644973
  0.3766823550724287  0.5053097168622148  0.5749873558237093
  0.6367691007874285  0.5526499384765567  0.5490239137818860
  0.6127892203220908  0.3937374206636431  0.5399676134016266
  0.4942310042139317  0.4983714104078595  0.4302292113427822
  0.5131211614858550  0.4951580522467757  0.4857880263515249
  0.5161012895884920  0.5481900662592318  0.4060527521349914
  0.5141783008547129  0.4503599148521812  0.4013841653162245
  0.4338197328645062  0.4995312045064069  0.4295491930790762


Zn7CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    20.4148643999999990    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.2386748999999995    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.9745029999999986
   Zn   C    H
     7     1     4
Direct
  0.5153613596216183  0.4857096804726639  0.6553559853340191
  0.3956275631884710  0.5807152831576943  0.4614028628367369
  0.4307868130120609  0.4584169179910119  0.5278862824433584
  0.5611434460306611  0.4016837240663813  0.5503699519900788
  0.6154480363882230  0.5366335996194945  0.5732323926378901
  0.4949146449450020  0.6044663429783229  0.5449062247929067
  0.3713223716631525  0.5735939520410869  0.5907873437208916
  0.5636172149857878  0.5184454256082330  0.4015315552161935
  0.5220109605823297  0.4866405279404750  0.4200603813227663
  0.5807702525227908  0.4942279429294451  0.3566071617917230
  0.6033960663217739  0.5173770299159752  0.4368048948780612
  0.5478712707381262  0.5752395732792098  0.3934349630353745


Zn8CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    20.3127001000000007    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.8111634999999993    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.6945778000000011
   Zn   C    H
```

```
     8     1     4
Direct
  0.4497205121626583  0.3786082986481716  0.5168900999608188
  0.6302234197120284  0.5616925697618074  0.5275790691583302
  0.3883878566079836  0.6228219006388958  0.5210534716911268
  0.5805765375373757  0.5283651439939290  0.6495648197309203
  0.5056691177350303  0.5893877739556089  0.5619867108569776
  0.4449231531761334  0.4499814857531043  0.6279500808113448
  0.3956397786870425  0.4945063532841990  0.5108359440992249
  0.5471452876486029  0.4612432994851256  0.5332806694615226
  0.4957110785529962  0.5657098133529390  0.3817986398166927
  0.4754424387686243  0.5208870328733568  0.4015683688927247
  0.5492245617343890  0.5607685833570911  0.3759797051114181
  0.4729253817875390  0.5761041160371023  0.3348605220796939
  0.4856508758895935  0.6054336288586666  0.4155018983292011


Zn9CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    19.7680031000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.5511237999999992    0.0000000000000000
     0.0000000000000000    0.0000000000000000   22.2692017000000000
   Zn   C    H
     9     1     4
Direct
  0.5285287403478868  0.4675047029018903  0.4677573642680826
  0.6304162985418099  0.5002860521114825  0.5416927712104941
  0.5352904317033674  0.5913039252131542  0.5204643268191320
  0.4281310738826677  0.4700881096599934  0.6253060044324681
  0.4098824832751027  0.5285361994116915  0.5091288530658981
  0.4289969738427752  0.4003891695699737  0.5224690288768034
  0.4406504240437581  0.5991859730701572  0.6075384309858606
  0.5466270341432691  0.4071810445583335  0.5837036200945463
  0.5535741207602016  0.5330687485887079  0.6365072684611642
  0.4380660996854469  0.5863451626648076  0.3458413115648439
  0.4667716881031556  0.5444667664885915  0.3289811984867743
  0.4668277541552802  0.6313192828572479  0.3384624654236554
  0.4289164644673589  0.5802259309258323  0.3942735826232142
  0.3892704130479333  0.5894589319781316  0.3224137736870715


Zn10CH4 shown in Figure S11
1:P1(1)
   1.00000000000000
    18.3920512000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.7483441999999982    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.2463696000000013
   Zn   C    H
    10     1     4
Direct
  0.4872480269244899  0.5231182490641868  0.5063205850916113
  0.4434535928162430  0.3841826018335978  0.4726587809190964
  0.6369948261020233  0.5356163492858323  0.4836007616419514
  0.5701224978831493  0.4255482111746861  0.4379336334361231
```

```
  0.4128235660504040  0.6200278576730609  0.4277507109055997
  0.4784155278901806  0.5494892390980241  0.2918337974966723
  0.4947360039520717  0.4120680522907892  0.3201794063239687
  0.4023638849012704  0.4882913974013965  0.3959233835669883
  0.6077256511126800  0.4995720255593544  0.3252381872286734
  0.5483687391041614  0.6008042097191906  0.4031462846510933
  0.4911183008310654  0.4856300347034494  0.6735475711585360
  0.4494459116397743  0.5161808605284677  0.6453745022807976
  0.5332961108702791  0.4678672572457068  0.6371093755647943
  0.4655280773993490  0.4388708687373186  0.6977404439706236
  0.5155792825228569  0.5196627856849512  0.7135825757634667


Zn1 shown in Figure S12
Zn
   1.00000000000000
    20.0000000000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Zn
     1
Selective dynamics
Direct
  0.5000000000000000  0.5000000000000000  0.5000000000000000   T   T   T


Zn2 shown in Figure S12
Zn
   1.00000000000000
    22.3859999999999992    0.0000000000000000    0.0000000000000000
     0.0000000000000000   20.0000000000000000    0.0000000000000000
     0.0000000000000000    0.0000000000000000   20.0000000000000000
   Zn
     2
Selective dynamics
Direct
  0.4289522515326422  0.4999999996550238  0.5000003852991850   T   T   T
  0.5710477484673578  0.5000000003449763  0.4999996147008151   T   T   T


Zn3 shown in Figure S12
1:P1(1)
   1.00000000000000
    16.5716861000000009    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.3371600999999984    0.0000000000000000
     0.0000000000000000    0.0000000000000000   15.0000000000000000
   Zn
     3
Direct
  0.5425933277314546  0.6001896677788473  0.4999657615437613
  0.4006292376404300  0.4763555898691583  0.5000265227573586
  0.5779874346281183  0.4137947423519909  0.5000077156988799


Zn4 shown in Figure S12
```

```
2:P1(1)
   1.00000000000000
    16.1741394000000014    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.7061168000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.3925009000000017
   Zn
     4
Direct
  0.5622692347630051  0.5000178766063494  0.4142784404805067
  0.5622888581236619  0.4999923109743844  0.5857571977283235
  0.4377020037804862  0.5830888652670686  0.4999992090464052
  0.4377499033328429  0.4169009471521976  0.4999651527447647


Zn5 shown in Figure S12
1:P1(1)
   1.00000000000000
    17.0428427999999990    0.0000000000000000    0.0000000000000000
     0.0000000000000000   16.1794012000000009    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.3244271000000012
   Zn
     5
Direct
  0.5604216766472352  0.6048068533243215  0.4999974820809847
  0.5108130248871559  0.5226984578856233  0.6455200706260048
  0.5108168932488942  0.5226847922338875  0.3544734632874011
  0.4186138668264009  0.5229927272812085  0.4999874025324568
  0.5592645383903081  0.4361571692749552  0.5000115814731565


Zn6 shown in Figure S12
2:P1(1)
   1.00000000000000
    17.9816935999999998    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.4841145000000004    0.0000000000000000
     0.0000000000000000    0.0000000000000000   16.4598402999999998
   Zn
     6
Direct
  0.4783419223200159  0.3791166415686840  0.5374371010495121
  0.5284268583647077  0.6535006822363774  0.5067641717695824
  0.5939993733070311  0.5135492927167796  0.5638458539206695
  0.4284656044190142  0.5278881854849073  0.5686226953713770
  0.3417642467585902  0.4304759725052545  0.4455819181485789
  0.4928919948306458  0.5067892254880020  0.4221982597402776


Zn7 shown in Figure S12
1:P1(1)
   1.00000000000000
    18.4196181999999986    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.0456056999999994    0.0000000000000000
     0.0000000000000000    0.0000000000000000   18.4774285999999996
   Zn
     7
```

```
Direct
  0.3538974758566713  0.5448407285260569  0.5209595486066878
  0.4913581838476551  0.5222541407229694  0.4300030350637972
  0.5005648619842191  0.4968452501044419  0.5717817307302178
  0.5890786438957157  0.3954906043559168  0.4728255318829804
  0.4866132869606122  0.6567525308177470  0.5277832023410994
  0.6320522774823801  0.5658985169468959  0.5000307000539402
  0.4161052699727470  0.3846182285259694  0.4864662513212774


Zn8 shown in Figure S12
1:P1(1)
   1.00000000000000
    19.8657414999999986    0.0000000000000000    0.0000000000000000
     0.0000000000000000   18.8458010000000016    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.5161763000000015
   Zn
     8
Direct
  0.4833038151587820  0.5056703158047449  0.6170126773453500
  0.4972983634458286  0.4659186075296343  0.3714677236586701
  0.5525340698912148  0.5459826659651529  0.5025905720952758
  0.4390515775953734  0.6150777128456851  0.5344723404241454
  0.4090938561110055  0.4804496295235872  0.5021827451778776
  0.3983777625119096  0.3702071068942801  0.4159471261668462
  0.5325309753877024  0.3827207087046541  0.4763161222140764
  0.6315195798981935  0.4768832527322646  0.4151606929177570


Zn9 shown in Figure S12
1:P1(1)
   1.00000000000000
    18.3542904000000000    0.0000000000000000    0.0000000000000000
     0.0000000000000000   19.1347214000000001    0.0000000000000000
     0.0000000000000000    0.0000000000000000   19.8780530000000013
   Zn
     9
Direct
  0.4551071438924084  0.6266682411140589  0.5231509016392882
  0.4901726343393436  0.4008133958212932  0.5709615630597011
  0.3876212726394808  0.4995051423398249  0.5531822919367074
  0.5973925359976522  0.4653108782367984  0.5023247757422004
  0.5987213011824500  0.6033000793093963  0.5228783994071605
  0.3887529364650750  0.5514896001256523  0.4276703145725961
  0.5340650903515018  0.5590991331551579  0.4092310271964190
  0.5203987537493893  0.5322244079951345  0.6149386287674116
  0.4649383313826935  0.4322891219026873  0.4401920976785135


Zn10 shown in Figure S12
1:P1(1)
   1.00000000000000
    17.9626458999999983    0.0000000000000000    0.0000000000000000
     0.0000000000000000   17.9524493000000014    0.0000000000000000
     0.0000000000000000    0.0000000000000000   17.8449598999999992
```

```
   Zn
    10
Direct
  0.4070228820163879  0.5419563499787345  0.5062309646720814
  0.4999845628527297  0.3784680045395823  0.5059084364716284
  0.5929849583698560  0.5419650530523387  0.5062332743074808
  0.5000061038552175  0.4862496760615311  0.3981099947112949
  0.4999943373727909  0.5714599674394946  0.6189114094578424
  0.4247539900895797  0.4433087886574267  0.6170610640827952
  0.5752499308133076  0.4433000109122063  0.6170498186835163
  0.6277252031788938  0.4140429390512239  0.4523256294486268
  0.5000061235143218  0.6347141985474769  0.4508241949534664
  0.3722719079369076  0.4140550117599934  0.4522952132112655
```

S12. Supplementary references

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(11) Since the $2p_z$ orbital of the C atom of CH_3 does not interact with the 1s orbitals of the three H's, the energy level of the PDOS peak for the $2p_z$ orbital of CH_3 is likely to be little changed from the energy level of the 2p orbitals of the gas-phase carbon atom.