TABLE I. Composition of the active space with 76 orbitals for FeMoco.

group	orbital	orbital index
		left cubane
1	left end	1, 2
2	Fe1 3d	3, 4, 5, 6, 7
3	S 3p	8, 9, 10, 11, 12, 13, 14, 15, 16
4	Fe2 3d	17, 18, 19, 20, 21
5	Fe3 3d	22, 23, 24, 25, 26
6	Fe4 3d	27, 28, 29, 30, 31
		central part
7	S 3p, C 2s2p	32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44
		right cubane
8	Fe5 3d	45, 46, 47, 48, 49
9	Fe6 3d	50, 51, 52, 53, 54
10	Fe7 3d	55, 56, 57, 58, 59
11	S 3p	60, 61, 62, 63, 64, 65, 66, 67, 68
12	Mo8 4d	69, 70, 71, 72, 73
13	right end	74, 75, 76

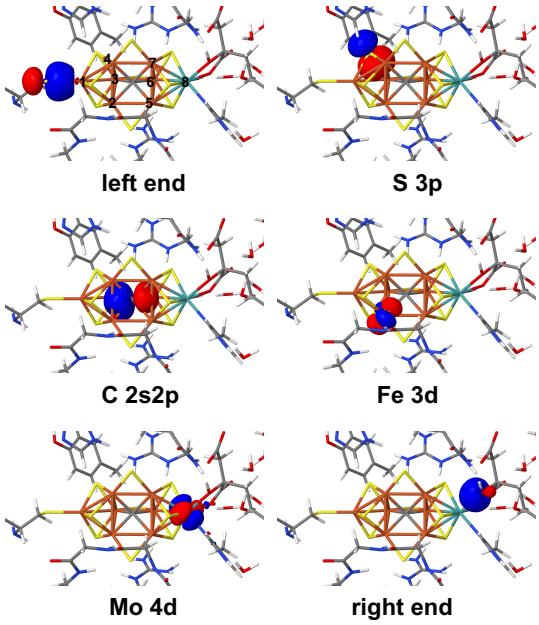

FIG. 3. Illustration of some selected active orbitals for FeMoco in the active space model CAS(113e,76o) constructed in this work.

[5]T. Spatzal, M. Aksoyoglu, L. Zhang, S. L. Andrade, E. Schleicher, S. Weber, D. C. Rees, and O. Einsle, Science **334**, 940 (2011).
[6]K. M. Lancaster, M. Roemelt, P. Ettenhuber, Y. Hu, M. W. Ribbe, F. Neese, U. Bergmann, and S. DeBeer, Science **334**, 974 (2011).
[7]M. Reiher, N. Wiebe, K. M. Svore, D. Wecker, and M. Troyer, Proc. Nat. Acad. Sci. **18**, 7555 (2017).
[8]I. Shavitt and R. J. Bartlett, *Many-body methods in chemistry and physics: MBPT and coupled-cluster theory* (Cambridge university press, 2009).
[9]S. R. White and R. L. Martin, J. Chem. Phys. **110**, 4127 (1999).
[10]G. K.-L. Chan and M. Head-Gordon, J. Chem. Phys. **116**, 4462 (2002).
[11]G. K.-L. Chan and S. Sharma, Annu. Rev. Phys. Chem. **62**, 465 (2011).
[12]S. Sharma and G. K.-L. Chan, J. Chem. Phys. **136** (2012), 10.1063/1.3695642.
[13]U. Schollwöck, Rev. Mod. Phys. **77**, 259 (2005).
[14]A. A. Holmes, N. M. Tubman, and C. J. Umrigar, J. Chem. Theory Comput. **12**, 3674 (2016).
[15]S. Sharma, A. A. Holmes, G. Jeanmairet, A. Alavi, and C. J. Umrigar, J. Chem. Theory Comput. **13**, 1595 (2017).
[16]J. Li, M. Otten, A. A. Holmes, S. Sharma, and C. J. Umrigar, Https://arxiv.org/pdf/1809.04600.
[17]E. Münck, H. Rhodes, W. Orme-Johnson, L. Davis, W. Brill, and V. Shah, Biochimica et Biophysica Acta (BBA)-Protein Structure **400**, 32 (1975).
[18]R. Zimmermann, E. Münck, W. J. Brill, V. K. Shah, M. T. Henzl, J. Rawlings, and W. H. Orme-Johnson, Biochimica et Biophysica Acta (BBA)-Protein Structure **537**, 185 (1978).
[19]S. J. Yoo, H. C. Angove, V. Papaefthymiou, B. K. Burgess, and E. Münck, Journal of the American Chemical Society **122**, 4926 (2000).
[20]R. Bjornsson, F. A. Lima, T. Spatzal, T. Weyhermüller, P. Glatzel, E. Bill, O. Einsle, F. Neese, and S. DeBeer, Chemical Science **5**, 3096 (2014).
[21]T. Spatzal, J. Schlesier, E.-M. Burger, D. Sippel, L. Zhang, S. L. Andrade, D. C. Rees, and O. Einsle, Nature communications **7**, 10902 (2016).
[22]R. Bjornsson, F. Neese, and S. DeBeer, Inorganic chemistry **56**, 1470 (2017).
[23]S. Sharma, K. Sivalingam, F. Neese, and G. K.-L. Chan, Nature Chem. **6**, 927 (2014).
[24]Z. Li and G. K.-L. Chan, Journal of Chemical Theory and Computation **13**, 2681 (2017).
[25]N. M. Tubman, C. Mejuto-Zaera, J. M. Epstein, D. Hait, D. S. Levine, W. Huggins, Z. Jiang, J. R. McClean, R. Babbush, M. Head-Gordon, and K. B. Whaley, Https://arxiv.org/pdf/1809.05523.pdf.
[26]Https://github.com/zhendongli2008/Active-space-model-for-FeMoco.
[27]A. D. Becke, The Journal of chemical physics **98**, 5648 (1993).
[28]C. Lee, W. Yang, and R. G. Parr, Physical review B **37**, 785 (1988).
[29]P. Stephens, F. Devlin, C. Chabalowski, and M. J. Frisch, The Journal of Physical Chemistry **98**, 11623 (1994).
[30]F. Jorge, N. A. Canal, G. Camiletti, and S. Machado, J. Chem. Phys. **130**, 064108 (2009).
[31]F. Weigend and R. Ahlrichs, Physical Chemistry Chemical Physics **7**, 3297 (2005).