

Theodore A. Betley, Ph.D.

EDUCATION

- 2007 **Postdoctoral Fellow, Chemistry**
Massachusetts Institute of Technology, Cambridge, MA
Advisor: Prof. Daniel G. Nocera
- 2005 **Ph.D., Chemistry**
California Institute of Technology, Pasadena, CA
Thesis Title: *Coordination chemistry at trigonally coordinated iron platforms: chemistry related to dinitrogen reduction*
Advisor: Prof. Jonas C. Peters
- 1999 **B.S.E. Chemical Engineering**, summa cum laude
University of Michigan, Ann Arbor, MI

EMPLOYMENT

- 2019 – present **Harvard University**, Director of Graduate Studies, Department Chair
- 2019 – present **Harvard University**, Erving Professor of Chemistry
- 2014 – 2019 **Harvard University**, Professor
- 2011 – 2013 **Harvard University**, Thomas D. Cabot Associate Professor
- 2007 – 2011 **Harvard University**, Assistant Professor
- 2005 – 2007 **Massachusetts Institute of Technology**, NIH NRSA Postdoctoral Fellow
- 2000 – 2005 **California Institute of Technology**

AWARDS

Kavli Emerging Leader in Chemistry Lecturer, ACS (2015). Presidential Early Career Award in Science and Engineering, DOE (2013). Henry and Camille Dreyfus Teacher Scholar Award (2013). National Academy of Sciences Award for Initiatives in Research in Area of Catalysis (2013). National Fresenius Award, Phi Lambda Epsilon (2013). DOE Early Career Research Program (2012-2017). George W. Merck Fellowship, Harvard University (2011). AFOSR Young Investigator Program (2011). NSF CAREER Award (2010). Milton Fund Award (2008). Dreyfus Foundation Fellowship in Environmental Chemistry (2008). MIT Technology Review TR35 (2008). NIH NRSA Postdoctoral Fellowship, MIT (2005). Division of Inorganic Chemistry Young Investigator Award, ACS (2005). Herbert Newby McCoy Award, California Institute of Technology (2005). DOD Pre-doctoral Fellowship (2000).

AREAS OF INTEREST

My lab is active in ligand design and ascertaining the resulting metal-based properties to elucidate both structure/function and electronic structure/function relationships. We have developed base-metal catalyzed C–H bond functionalization through our understanding of the electronic structure underpinnings to developing reactive metal-ligand multiple bonds. The desirable function of these complexes to undergo atom- or functional group transfer into unactivated C–H bonds is driven by destabilizing the catalyst. Destabilizing the catalyst is achieved by targeting high-spin or open-shell formulations. Population of metal-ligand antibonding orbitals diminishes metal-ligand covalency, leading to greater ligand lability. Furthermore, high-spin metal centers can stabilize ligand-based radicals, giving rise to highly reactive ligand functionalities primed to activate C–H bonds. We have developed dipyrin coordination complexes featuring first row transition metals to facilitate formation of the high-spin metal ligand multiple bonds. Targeting high-spin metal complexes has allowed us to achieve catalytic intermolecular and intramolecular C–H amination; render imido complexes reactive for C–H bond activation where their low-spin analogues are inert; and use ancillary ligand tuning to stabilize ligand-based radicals and render cyclization reactions diastereoselective. We have developed our understanding of reaction site design to include polynuclear reaction sites. Our group's primary innovation derives from our ability to synthesize reactive polynuclear

complexes that can functionally, and electronically, model cluster cofactors from nature. We have developed synthetic methodology permitting the isolation of substitutionally homogeneous, homo- and hetero-trinuclear clusters. We are able to characterize these materials by a suite of spectroscopic techniques to make detailed connections between cluster composition, its resulting electronic structures, and ensuing reactivity. We have developed a new class of materials that we propose are, from an electronic structure standpoint, representative of the critical design features of clusters found in metalloenzymes. Given this connection we are applying a variety of spectroscopic techniques to understand the influence of M–M interactions within cluster aggregates to perform redox, delocalize oxidative stress, and perform small molecule activation processes. This combination of synthesis, spectroscopy, and electronic structure determination allows us to demonstrate how synthetic clusters, and by extension, propose how clusters in biology perform a variety of functions. My contributions to the community beyond science include: serving on the Editorial Advisory Boards of *Chemical Society Reviews*, *Chem*; Co-PI for Crystallography Line at ChemMatCARS at the Advanced Photon Source, Argonne National Lab.

PUBLICATIONS

1. Dong, Y.; Wrobel, A. T.; Porter, G. J.; Kim, J. J.; Essman, J.; Zheng, S.-L.; Betley, T. A. "O–Heterocycle synthesis via intramolecular C–H alkoxylation catalyzed by iron acetylacetonate." *J. Am. Chem. Soc.* **2021**, *143*, 7480-7489.
2. Carsch, K. M.; Iliescu, A.; McGillicuddy, R. D.; Mason, J. A.; Betley, T. A. "Reversible scavenging of dioxygen from air by a copper complex." *J. Am. Chem. Soc.* (*in revision*).
3. Nehrkorn, J.; Greer, S. M.; Malbrecht, B.; Anderton, K.; Aliabadi, A.; Krzystek, J.; Schnegg, A.; Holldack, K.; Herrmann, C.; Betley, T. A.; Stoll, S.; Hill, S. "Spectroscopic investigation of a metal-metal bonded Fe₆ single-molecule magnet with an isolated $S = 19/2$ giant-spin ground state." *Inorg. Chem.* **2021**, *60*, 4610-4622.
4. Bartholomew, A. K.; Musgrave, R. A.; Anderton, K. J.; Juda, C. E.; Dong, Y.; Bu, W.; Wang, S.-Y.; Chen, Y.-S.; Betley, T. A. "Revealing redox isomerism in trichromium imides by anomalous diffraction." *Submitted*.
5. Juda, C. E.; Bartholomew, A. K.; Powers, T. M.; Gu, N. X.; Meyer, E.; Roth, N.; Chen, Y.-S.; Zheng, S.-L.; Betley, T. A. "Cluster dynamics of heterometallic trinuclear clusters during reactivity and redox processes." *Submitted*.
6. Johnson, E. J., Betley, T. A. "Redox-triggered hydroxy release from a dicobalt bis-hydroxo complex." *Submitted*.
7. Scharf, A. B., Zheng, S.-L., Betley, T. A. "Luminescence from open-shell, first-row transition metal dipyrin complexes." *Dalton Trans.* **2021**, *50*, 6418-6422.
8. Carsch, K. M.; Lukens, J. T.; Elder, S. E.; Dogutan, D. K.; Nocera, D. G.; Yang, J.; Zheng, S.-L.; Daniel, T.; Betley, T. A. "Synthesis and solid-state structures of two cofacial (bis)dipyrin dichromium complexes in different charge states." *Acta Cryst.*, **2021**, *C77*, 161-166.
9. Dong, Y. Lund, C. J.; Porter, G. J.; Clarke, R. M.; Zheng, S.-L.; Cundari, T. R.; Betley, T. A. "Enantioselective C–H amination catalyzed by nickel iminyl complexes supported by anionic bisoxazoline (BOX) ligands." *J. Am. Chem. Soc.* **2021**, *143*, 817-829.
10. Dong, Y.; Clarke, R. M.; Porter, G. J.; Betley, T. A. "Efficient C–H amination catalysis using nickel-dipyrin complexes." *J. Am. Chem. Soc.* **2020**, *142*, 10996-11005.
11. Teesdale, J. J.; Kwak, J.; Su, A. C.; Caputo, C. B.; Crockett, M. P.; Chiaie, K. R. D.; Betley, T. A. "The dramatic impact of single atom substitutions: cathodic shifting and electronic structure changes in isovalent clusters." *Submitted*.
12. Teesdale, J. J.; Greer, S. M.; Hill, S.; Betley, T. A. "Conferring magnetic anisotropy to high spin clusters." *Submitted*.
13. Wrobel, A. T.; Lukens, J. T.; Lancaster, K. M.; Betley, T. A. "Iron stabilized high spin carbenes." *Submitted*.
14. Dong, Y.; Clarke, R. M.; Zheng, S.-L.; Betley, T. A. "Synthesis and electronic structure studies of a Cr-imido redox series." *Chem. Commun.* **2020**, *56*, 3163-3166.

15. Baek, Y.; Betley, T. A. "Reversible C–C bond cleavage of cobalt diketimide into elusive cobalt iminyl complex." *Angew. Chem. Int. Ed. (In revision)*.
16. Carsch, K. M.; Lukens, J. T.; DiMucci, I. M.; Iovan, D. A.; Zheng, S.-L.; Lancaster, K. M.; Betley, T. A. "Electronic structures and reactivity profiles of aryl nitrenoid-bridged dicopper complexes." *J. Am. Chem. Soc.* **2020**, *142*, 2264-2276.
17. Baek, Y.; Das, A.; Zheng, S.-L.; Powers, D. C.; Betley, T. A. "C–H amination mediated by cobalt organoazide adducts and the corresponding cobalt nitrenoid intermediates." *J. Am. Chem. Soc.* **2020**, *142*, 11232-11243.
18. Dong, Y.; Lukens, J. T.; Clarke, R. M.; Zheng, S.-L.; Lancaster, K. M.; Betley, T. A. "Synthesis, characterization and C–H amination of nickel iminyl complexes." *Chem. Sci.* **2020**, *11*, 1260-1268.
19. Baek, Y.; Hennessy, E. T.; Betley, T. A. "Direct manipulation of metal imido geometry: key principles to enhance C–H amination efficacy." *J. Am. Chem. Soc.* **2019**, *141*, 16944-16953.
20. DiMucci, I. M.; Lukens, J. T.; Chatterjee, S.; Carsch, K. M.; Titus, C. J.; Lee, S. J.; Nordlund, D.; Betley, T. A.; MacMillan, S. N.; Lancaster, K. M. "The myth of d⁸ copper(III)." *J. Am. Chem. Soc.* **2019**, *141*, 18508-18520.
21. Baek, Y.; Betley, T. A. "Catalytic C–H amination mediated by dipyrin cobalt imidos." *J. Am. Chem. Soc.*, **2019**, *141*, 7797-7806.
22. Carsch, K. M.; DiMucci, I.; Iovan, D. A.; Li, A.; Zheng, S.-L.; Titus, C. J.; Lee, S. J.; Irwin, K. D.; Nordlund, D.; Lancaster, K. M.; Betley, T. A. "Synthesis of a copper-supported triplet nitrene complex pertinent to copper-catalyzed amination." *Science* **2019**, *365*, 1138–1143.
23. Johnson, E. J.; Kleinlein, C.; Musgrave, R. A.; Betley, T. A. "Diiron oxo reactivity in a weak-field environment." *Chem. Sci.*, **2019**, *10*, 6304-6310.
24. Bartholomew, A. K.; Juda, C. E.; Lin, B.; Betley, T. A. "Ligand-based control of single-site vs multi-site reactivity by a trichromium cluster." *Angew. Chem. Int. Ed.*, **2019**, *58*, 5687-5691.
25. Bartholomew, A. K.; Teesdale, J. J.; Hernández Sánchez, R.; Malbrecht, B.; Juda, C. E.; Ménard, G.; Bu, W.; Iovan, D. A.; Mikhailine, A. A.; Zheng, S.-L.; Sarangi, R.; Wang, S. G.; Chen, Y.-S.; Betley, T. A. "Exposing the inadequacy of redox formalisms by resolving redox inequivalence within isovalent clusters." *Proc. Nat. Acad. Sci.*, **2019**, *116*, 15836-15841.
26. Hernández Sánchez, R.; Betley, T. A. "Thermally persistent high spin ground states in octahedral iron clusters." *J. Am. Chem. Soc.* **2018**, *140*, 16792-16806.
27. Mikhailine, A. A.; Kleinlein, C.; Zheng, S.-L.; Betley, T. A. "Iron-catalyzed synthesis of unprotected, complex N-heterocycles via direct amination of aliphatic C–H bonds." *J. Am. Chem. Soc.*, *Submitted*.
28. Iovan, D. A.; Wilding, M. J. T.; Baek, Y.; Hennessy, E. T.; Betley, T. A. "Diastereoselective C–H bond amination for disubstituted pyrrolidines." *Angew. Chem. Int. Ed.* **2017**, *56*, 15599-15602.
29. Wilding, M. J. T.; Iovan, D. A.; Wrobel, A. T.; Luken, J. T.; MacMillan, S. N.; Lancaster, K. M.; Betley, T. A. "Direct comparison of C–H bond amination efficacy through manipulation of nitrogen-valance centered redox: imido versus iminyl." *J. Am. Chem. Soc.* **2017**, *139*, 14757-14766.
30. Iovan, D. A.; Wrobel, A. T.; McClelland, A. A.; Scharf, A. B.; **Edouard, G. A.**; Betley, T. A. "Reactivity of a stable copper-dioxygen complex." *Chem. Commun.* **2017**, *53*, 10306-10309.
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33. Kleinlein, C.; Zheng, S.-L.; Betley, T. A. "Ground state and excited state tuning in ferric dipyrin complexes promoted by ancillary ligand exchange." *Inorg. Chem.* **2017**, *56*, 5892-5901.
34. Hernández Sánchez, R.; Bartholomew, A. K.; Powers, T. A.; Menard, G.; Betley, T. A. "Maximizing electron exchange in a [Fe₃] cluster." *J. Am. Chem. Soc.* **2016**, *138*, 2235-2243.
35. Iovan, D. A.; Betley, T. A. "Characterization of iron-imido species relevant for N-group transfer chemistry." *J. Am. Chem. Soc.* **2016**, *138*, 1983-1993.

36. Powers, T. M.; Bartholomew, A. M.; Juda, C. E.; Kwak, J.; Hernández Sánchez, R.; Betley, T. A. "Cooperative and site-isolated activation of π -acidic substrates on a high spin, triiron cluster." *Submitted*.
37. Hernández Sánchez, R.; Betley, T. A. "Meta-atom behavior in clusters revealing large spin ground states." *J. Am. Chem. Soc.* **2015**, *137*, 13949-13956.
38. Hernández Sánchez, R.; Zheng, S.-L.; Betley, T. A. "Ligand field strength mediates electron delocalization in octahedral $[(^H\text{L})_2\text{Fe}_6(\text{L}')_m]^{n+}$ clusters." *J. Am. Chem. Soc.* **2015**, *137*, 11126-11143.
39. Hernández Sánchez, R.; Willis, A. M.*; Zheng, S.-L.; Betley, T. A. "Synthesis of well-defined bicapped-octahedral iron clusters $[(^{\text{trcn}}\text{L})_2\text{Fe}_8(\text{PMe}_2\text{Ph})_2]^n$ ($n = 0, -1$)." *Angew. Chem., Int. Ed.* **2015**, *54*, 12009-12013.
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41. Sazama, G. T.; Betley, T. A. "Multiple, disparate redox pathways exhibited by a tris(pyrrolido)ethane iron complex." *Inorg. Chem.* **2014**, *53*, 269-281.
42. Powers, T. M.; Gu, N. X.*; Fout, A. R.; Baldwin, A. M.*; Hernández Sánchez, R.; Alfonso, D. M.; Chen, Y.-C.; Zheng, S.-L.; Betley, T. A. "Synthesis of open-shell, bimetallic Mn/Fe trinuclear clusters." *J. Am. Chem. Soc.* **2013**, *135*, 14448-14458.
43. Powers, T. M.; Betley, T. A. "Testing the polynuclear hypothesis: multi-electron reduction of small molecules by triiron reaction sites." *J. Am. Chem. Soc.* **2013**, *135*, 12289-12296.
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45. Hennessy, E. T.; Betley, T. A. "Complex *N*-heterocycle synthesis via iron-catalyzed, direct C–H bond amination." *Science* **2013**, *340*, 591-595.
46. Eames, E. V.; Hernández Sánchez, R.; Betley, T. A.; "Metal atom lability in polynuclear complexes." *Inorg. Chem.* **2013**, *52*, 5006-5012.
47. King, E. R.; Sazama, G. T.; Betley, T. A. "Co(III) imidos exhibiting spin crossover and C–H bond activation." *J. Am. Chem. Soc.* **2012**, *134*, 17858-17861.
48. Fout, A. R.; Xiao, D. J.*; Zhao, Q.; Harris, T. D.; King, E. R.; Eames, E. V.; Zheng, S.-L.; Betley, T. A.; "Trigonal Mn_3 and Co_3 clusters supported by weak-field ligands: a structural, spectroscopic, magnetic, and computational investigation into the correlation of molecular and electronic structure." *Inorg. Chem.* **2012**, *51*, 10290-10299.
49. Eames, E. V.; Betley, T. A.; "Site-isolated redox reactivity in a trinuclear iron complex." *Inorg. Chem.* **2012**, *51*, 10274-10278.
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51. Fout, A. R.; Zhao, Q.; Xiao, D. J.*; Betley, T. A. "Oxidative atom-transfer to a trimanganese complex to form $\text{Mn}_6(\mu^6\text{-E})$ ($\text{E} = \text{O}, \text{N}$) clusters featuring interstitial oxide and nitride functionalities." *J. Am. Chem. Soc.* **2011**, *133*, 16750-16753.
52. Harris, T. D.; Betley, T. A. "Multi-site reactivity: reduction of six equivalents of nitrite to give an $\text{Fe}_6(\text{NO})_6$ cluster with a dramatically expanded octahedral core." *J. Am. Chem. Soc.* **2011**, *133*, 13852-13855.
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54. Scharf, A. B.; Betley, T. A. "Electronic perturbations of iron dipyrinato complexes via ligand β -halogenation and meso-fluoroarylation." *Inorg. Chem.* **2011**, *50*, 6837-6845.
55. Harris, T. David; Zhao, Q.; Hernández Sánchez, R.; Betley, T. A. "Expanded redox accessibility via ligand substitution in the octahedral clusters Fe_6Br_6 cluster." *Chem. Commun.* **2011**, *47*, 6344-6347.
56. Zhao, Q.; Harris, T. D.; Betley, T. A.; " $[(^H\text{L})_2\text{Fe}_6(\text{NCMe})_m]^{n+}$ ($m = 0, 2, 4, 6; n = -1, 0, 1, 2, 3, 4, 6$): An electron-transfer series featuring octahedral Fe_6 clusters supported by a hexaamide ligand platform." *J. Am. Chem. Soc.* **2011**, *133*, 8293-8306.

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58. Powers, T. M.; Fout, A. R.; Zheng, S.-L.; Betley, T. A.; "Oxidative group transfer to a triiron complex, formation of a nucleophilic $[\text{Fe}_3(\mu^3\text{-N})]^-$." *J. Am. Chem. Soc.* **2011**, *133*, 336-338.
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PATENTS

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