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Metalloprotein Design: From Metal Binding to Control Catalytic Activity and Excited-state Dynamics

Proteins can function as evolvable multidentate ligands, imposing geometric and electronic constraints that tune metal-centered properties beyond those accessible in small-molecule systems. Conversely, metallocofactors can impart unique chemical and photophysical reactivities to protein matrices. These synergistic interactions between biochemical and inorganic components can now be systematically engineered through advances in data-driven protein design and genetic code expansion, enabling the incorporation of both canonical and noncanonical amino acids with tailored metal-chelating functionalities. In this work, we demonstrate precise control over the ligand environment, enabling the formation of unconventional coordination geometries. Protein scaffolds impose structural constraints that modulate excited-state lifetimes. In addition, minimal metallocofactors are identified as intrinsic, genetically encodable chromophores that enable light-activated protein modification. Overall, metalloprotein design provides a versatile platform to integrate catalysis and photochemistry, offering new opportunities in artificial metalloenzymes and photocatalysis.

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