Table S1. X-ray diffraction data collection and refinement statistics.

Crystal parameters	Ag^{+}-DzFer	Cu^{2+}-DzFer
Data Collection		
Beam line	SSRF-BL02U1	SSRF-BL18U1
Wavelength (Å)	0.979	0.979
Space group	I432	I432
Cell dimensions		
a, *b*, *c* (Å)	148.21, 148.21, 148.21	151.01, 151.01, 151.01
α, β, γ (°)	90, 90, 90	90, 90, 90
Resolution (Å)†	104.80-1.90 (2.00-1.90)	37.75-2.26 (2.34-2.26)
No. of observed reflections†	803649	453153
No. of unique reflections†	22119	14068
Completeness (%)†	100.0 (100.0)	99.9 (100.0)
Mean I/sigma(I)†	12.5 (2.9)	7.9 (0.6)
Multiplicity†	36.3 (35.3)	32.2 (33.1)
Wilson *B* factor (Å)	30.9	28.9
R_{merge}†	0.260 (1.752)	0.219 (0.595)
$CC_{1/2}$†	0.995 (0.924)	0.997 (0.988)
Refinement		
Program	PHENIX/REFMAC5	PHENIX/REFMAC5
R_{work}/R_{free}	0.151/0.190	0.164/0.226
Mean temperature factor (Å^2)	36.0	33.0
No. of non-H atoms	1585	1534
Protein	1375	1366
Water	193	155
Ligands	17	13
Average *B* value (Å^2)		
Protein	33.0	30.7
Water	43.9	39.0
Ligands	66.7	111.3
R.m.s. deviations		
Bond lengths (Å)	0.007	0.007
Bond angles (°)	0.847	0.811
Ramachandran plot, residues in		
Favoured region (%)	98.2	98.2
Allowed region (%)	1.8	1.8
Outlier region (%)	0.0	0.0
Clashscore	4.40	2.97
PDB code	8GY1	8HCT

†Values in parentheses correspond to the highest resolution shell.

Table S2. The percentage contents of secondary structure of DzFer at varying pH conditions (pH 2–12).

Sample	*α*-helix	*β*-sheet	*β*-turn	random coil
DzFer (pH 2)	62.37±0.80	22.3±2.4	10.57±1.40	4.73±0.50
DzFer (pH 4)	78.8±0.3	0	21.2±0.3	0
DzFer (pH 6)	82.5	0	17.5	0
DzFer (pH 8)	90.8±0.2	0	9.2±0.2	0
DzFer (pH 10)	88.67±0.10	0	11.33±0.10	0
DzFer (pH 12)	33.83±2.30	53.0±7.3	5.0±4.8	8.13±1.20

Table S3. The composition ratios of secondary structure of DzFer, Ag^{+}-DzFer, and Cu^{2+}-DzFer.

Sample	*α*-helix	*β*-sheet	*β*-turn	random coil
DzFer	90.8±0.2	0	9.2±0.2	0
Ag^{+}-DzFer	73.8±0.8	0	26.1±0.8	0
Cu^{2+}-DzFer	86.6±0.3	0	13.3±0.3	0

Table S4. The percentages of secondary structure of DzFer, Ag^{+}-DzFer, and Cu^{2+}-DzFer after Fe^{2+} ion uptake.

Sample	*α*-helix	*β*-sheet	*β*-turn	random coil
DzFer+Fe^{2+}	84.5±0.7	0	15.5±0.7	0
Ag^{+}-DzFer+Fe^{2+}	72.2±0.7	0	27.7±0.7	0
Cu^{2+}-DzFer+Fe^{2+}	85.7±0.4	0	14.3±0.4	0

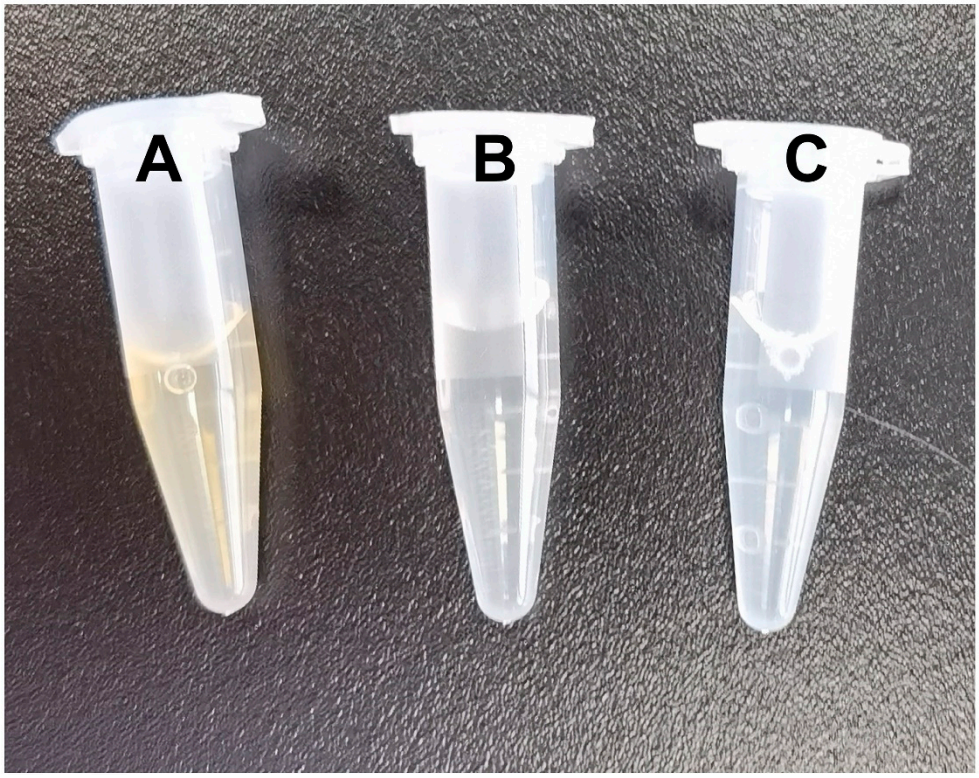


Figure S1. Solution images of DzFer with the concentrations of 5 mg/mL **(A)** and 1 mg/mL **(B)** as well as binding buffer (25 mM Tris–HCl pH 8.0, 150 mM NaCl) **(C)**.

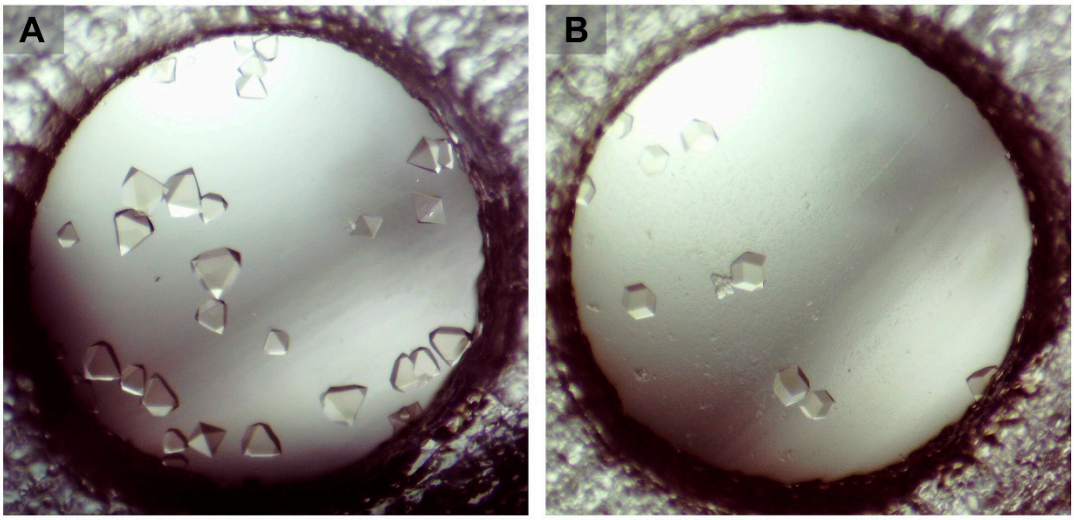


Figure S2. Crystals of DzFer **(A)** and Cu^{2+}-DzFer **(B)**.

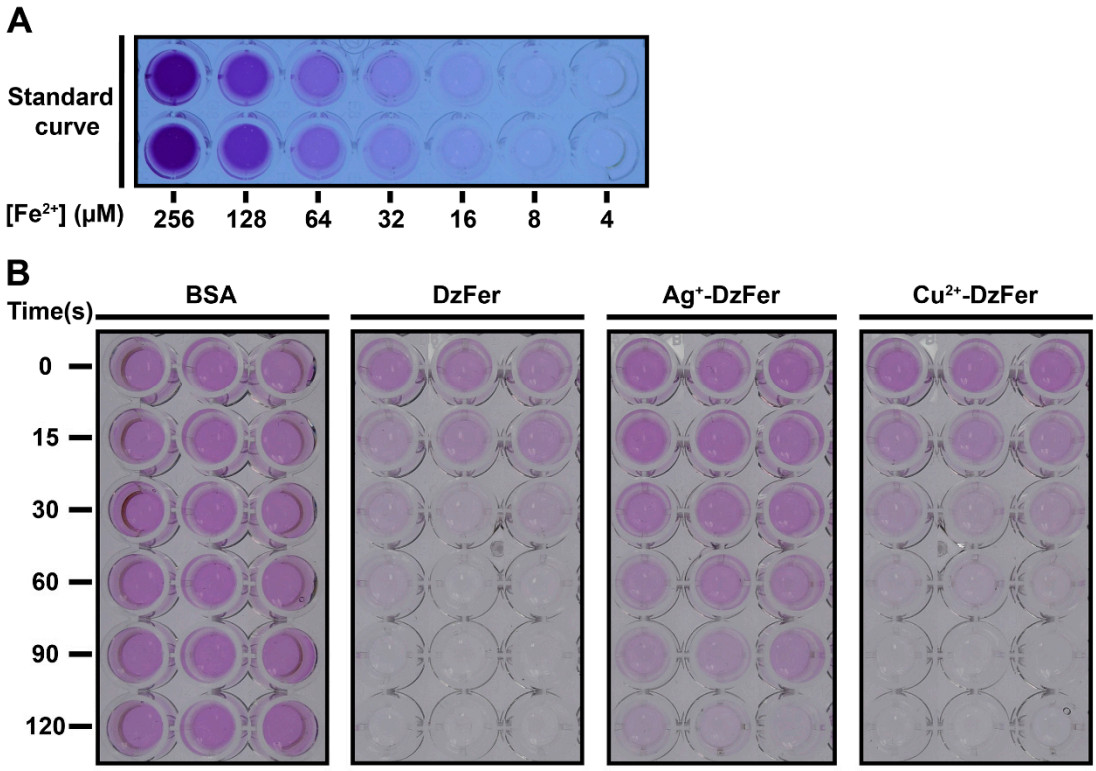


Figure S3. (A) A standard curve was drawn based on reading the optical density value at 560 nm by adding ferrozine into known serially diluted different Fe^{2+} concentrations; **(B)** The color reaction by monitoring the ferrozine–Fe^{2+} complexes at different time points revealed the protein samples oxidating Fe^{2+} into Fe^{3+}.

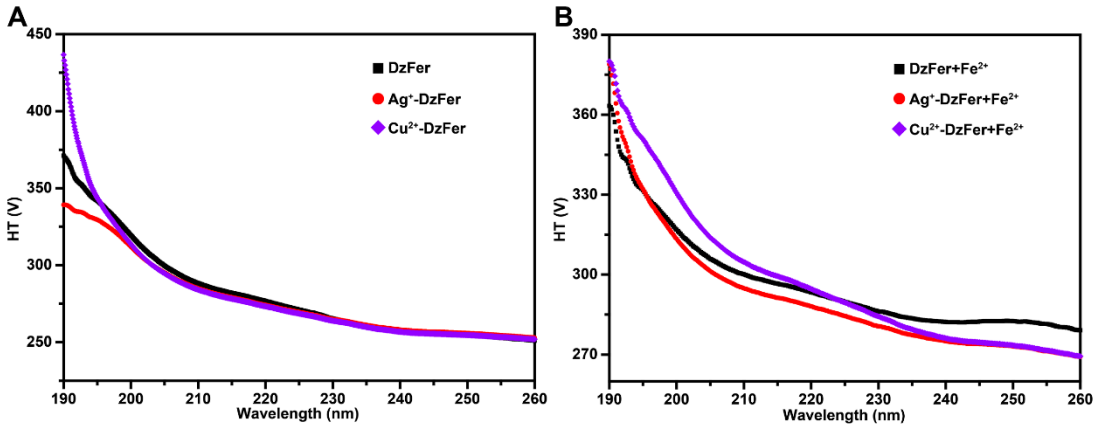


Figure S4. The recorded signals of the high-tension voltage (HT) during the CD measurement. **(A)** The HT traces of DzFer, Ag^{+}-DzFer, and Cu^{2+}-DzFer proteins; **(B)** The HT traces of DzFer+Fe^{2+}, Ag^{+}-DzFer+Fe^{2+}, and Cu^{2+}-DzFer+Fe^{2+} proteins.

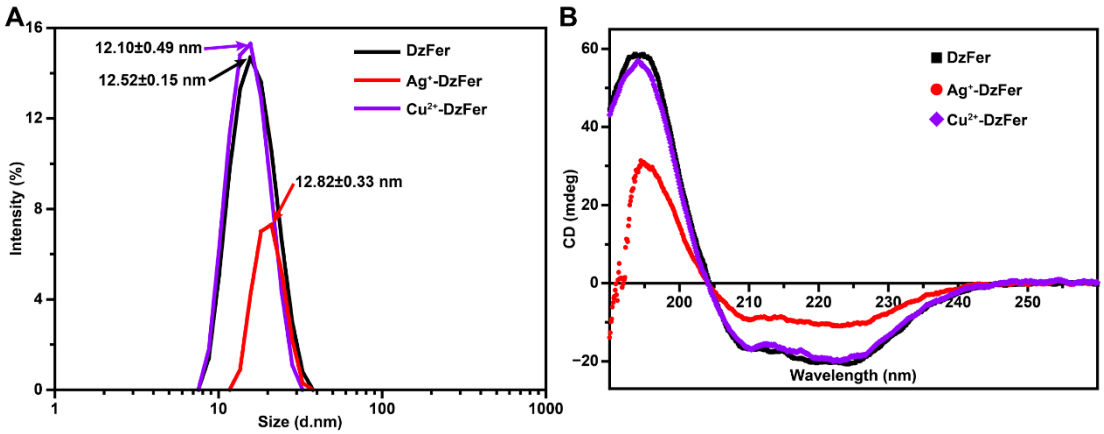


Figure S5. (A) DLS analysis of DzFer and metal-bound DzFer; **(B)** CD spectra of DzFer, Ag^{+}-DzFer, and Cu^{2+}-DzFer proteins.

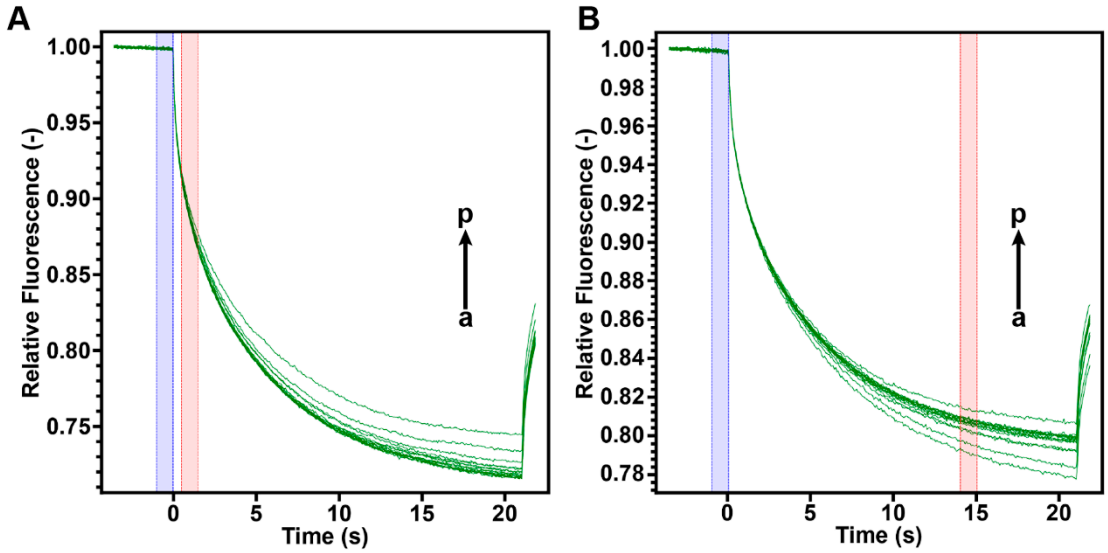
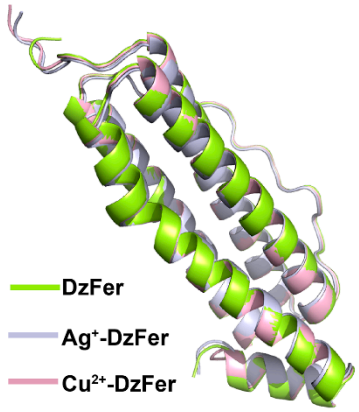


Figure S6. MST-measurements with DzFer and Ag^{+} or Cu^{2+}, respectively. **(A)** MST time traces of fluorescently labeled DzFer (approx. 200 nM) in the presence of different concentrations of Ag^{+} (from 0.275 nM to 9 μM, for curves a to p, respectively); **(B)** MST time traces of fluorescently labeled DzFer (approx. 200 nM) in the presence of different concentrations of Cu^{2+} (from 0.61 nM to 20 μM, for curves a to p, respectively). Scans were recorded at 20% excitation power at 25 °C.

A



B

	DzFer (168 residues)	Ag⁺-DzFer (169 residues)	Cu²⁺-DzFer (169 residues)
α-helix	75.6	75.1	74.6
β-sheet	0.0	0.0	0.0
β-turn	9.5	7.7	9.4
random coil	14.9	17.2	16.0

Figure S7. (A) The structural superposition of DzFer, Ag^{+}-DzFer, and Cu^{2+}-DzFer subunits; **(B)** The percentages of the secondary structure assignments based on the known crystal structures of DzFer (PDB ID: 7EMK), Ag^{+}-DzFer (PDB ID: 8GY1), and Cu^{2+}-DzFer (PDB ID: 8HCT).

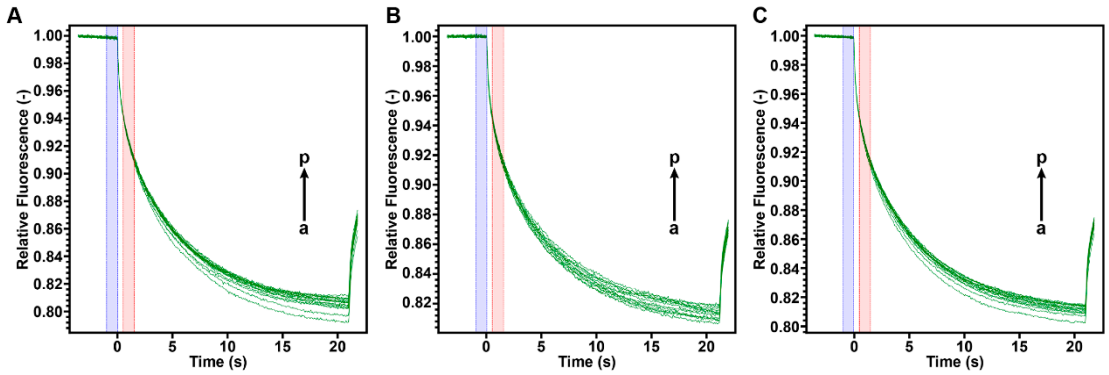


Figure S8. MST time traces of fluorescently labeled protein samples (approx. 200 nM), e.g., DzFer **(A)**, Ag^{+}-DzFer **(B)**, and Cu^{2+}-DzFer **(C)**, in the presence of different concentrations of Fe^{2+} (from 0.61 nM to 20 μM, for curves a to p, respectively). Scans were recorded at 20% excitation power at 25 °C.