In the homometallic carbido and nitrido clusters, **3** and **4**, the bond distance from the heteroatom to the hinge metal (M_h) atoms is considerably longer than the distance from the heteroatom to the wingtip metal (M_w) atoms (Table 1). Such a distinct bonding preference is not observed for **2**. Additionally, the elongation of the M_h–M_w distance relative to the M_h–M_h distance in **3** and **4** is not observed in **2**. The Fe–Fe distances in **2** are nearly unperturbed from those seen in **1** (C_{3v} symmetry).[3f] As a consequence of these unique structural features, the M_w–L–M_w angle in **2** is more acute than in the nitrido and carbido clusters, **3** and **4**, respectively, and the dihedral angle between the two wings of the butterfly in **2** is approximately 10° larger than in **3** and **4**.

Table 1. Structural parameters of μ_4-L butterfly clusters [a].

	2	3	4
M_h-L [Å]	1.902(4)	1.96(3)	1.900(6)
M_w-L [Å]	1.872(3), 1.877(3) [b]	1.80(3)	1.771(5)
M_w-M_h [Å]	2.517(2), 2.61(2) [b]	2.64(1)	2.604(7)
M_h-M_h [Å]	2.499(1)	2.533(2)	2.512(1)
M_w-L-M_w [°]	168.3(1)	176.3(4)	179.0(3)
M_w-M_h-M_h-M_w [°]	113	102	102
Ref.	this work	[10]	[11]

[a] M_w, wingtip metal atom; M_h, hinge metal atom. [b] For the first entry, M = Fe; for the second entry, M = Mn.

The oxo clusters **1** and **2** were characterized by ^{17}O-NMR spectroscopy in acetone solution at room temperature on samples enriched with [17]O at the oxo ligand. For **1**, the spectrum consists of a resonance for the CO ligands ($\delta = 356$) and one for the μ_3-oxo ligand ($\delta = 107$). For **2**, two distinct resonances are seen for the CO ligands ($\delta = 385$ and 370, intensity ratio 1:3) and the μ_4-oxo ligand resonance is found at $\delta = 93$. The relative positions of the oxo ligand resonances in **1** and **2** is in qualitative agreement with the correlation observed between the oxygen atom coordination number and the [17]O chemical shift of the oxygen atom in polyoxometalate systems.[12]

Two resonances for the CO ligands of **2** ($\delta = 214.0$; 225.5; intensity ratio 3:1) were also observed in the ^{13}C-NMR spectrum (CD$_2$Cl$_2$/CHFCl$_2$ (1:2), room temperature). These two resonances correspond to the carbonyl ligands bound to the Mn atom and those bound to the Fe atoms. A fluxional process involving equilibration of the hinge and wingtip Fe-bound CO ligands must be occurring at room temperature to account for this pattern. At -50°C, this equilibration process is not rapid and distinct resonances are seen for the two types of Fe(CO)$_3$ fragments (Mn(CO)$_3$, $\delta = 225.0$; Fe(CO)$_3$, $\delta = 212.1$ and 214.2; intensity ratio 3:3:6).

Unlike the nitrido[5a] and carbido[5b] butterfly clusters and the parent cluster **1**, **2** does not display ready reactivity toward Brønsted acids. Addition of one equivalent of CF$_3$SO$_3$H to a CH$_2$Cl$_2$ solution of **2** results in only negligible protonation.

The rarity of low-valent oxo clusters seems surprising given the vast chemistry that has been developed for the carbido and nitrido clusters. The apparently facile loss of an oxo ligand as CO$_2$ in carbonyl-containing complexes may, in part, offer an explanation. For instance, **1** decomposes under CO (1 atm) in acetone over a period of several hours to give CO$_2$ and lower-nuclearity Fe compounds; such a ready decomposition route is not accessible for ni-

trido and carbido clusters. If this route for loss of the oxo ligand can be suppressed, higher-nuclearity oxo clusters of low-oxidation-state metals should be accessible utilizing lower-nuclearity oxo clusters as precursors.

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[8] **2**: $a = 17.100(2)$, $b = 15.890(1)$, $c = 17.417(2)$ Å, $\beta = 94.90(1)$° at -120°C. Space group $P2_1/c$ ($Z = 4$). The full matrix refinement of 613 parameters with 5607 reflections converged at $R = 0.037$, $R_w = 0.044$, GOF = 1.77. Further details of the crystal structure investigation may be obtained from the Fachinformationszentrum Energie, Physik, Mathematik GmbH, D-7514 Eggenstein-Leopoldshafen 2 (FRG), on quoting the depository number CSD-52248, the names of the authors, and the journal citation.

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The Diradical 2,3,5,6-Tetramethylene-1,4-cyclohexanediyl ("1,2,4,5-Tetramethylenebenzene")**

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The title compound **10**, a disjoint diradical with conjugated non-Kekulé structure, is of topical interest in discussing the limits of the Hunds rule.[1] High-level ab initio SCF-MO-CI calculations lead one to expect a singlet ground state for **10** which lies about 5–7 kcal/mol below the lowest triplet state.[2] We have now checked this prediction experimentally.

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