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### **Spin Accommodation and Reactivity of Superatoms.<sup>1</sup>**

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We have recently discovered novel effects that may allow tuning of the reactivity of small clusters by controlling their spin excitation, electronic structure, and local geometry. These findings offer the prospect of designing novel catalysts through cluster assemblies where chosen clusters, called superatoms, serve as elemental building blocks. Taking aluminum as an example, I will present our recent findings that illustrate how reactive clusters can be made non-reactive while inert species can be made reactive by adding hydrogen atoms. These findings offer a microscopic understanding of the recent experimental reactivity studies on aluminum and aluminum-hydrogen clusters that show variable reactivity in even electron systems and rapid etching in odd electron systems. It is shown that the reactivity of even electron clusters is governed by a spin transfer, from the triplet oxygen to the cluster, that fills the spin down antibonding orbitals on oxygen. Theoretical investigations show that when the spin transfer cannot occur, the species is unreactive, and when spin accommodation is possible, more subtle effects appear. Secondly, I will examine the reactivity of aluminum clusters with simple nucleophiles such as water. The reactivity and nature of the ensuing products is wildly variable with the size and shape of the cluster. Again, the electronic structure and local coordination of the active sites allow for an understanding of changing barrier heights and resulting reactivity. This work provides a framework with which new catalysts may be designed.

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