Many challenging chemical reactions require precious metal catalysts to proceed. Our work utilizes group 13 metalloligands (Al, Ga, and In) as an electronic lever for tuning a reactive transition metal active site via a direct metal–metal interaction. This approach has allowed for the development and optimization of highly efficient metal–group 13 bifunctional catalysts for the hydrogenation of carbon dioxide. The bimetallic catalysts display remarkably enhanced activity compared to the analogous single metal centers. In this talk, I will detail the roles of the group 13 support in substrate binding, activation, and catalysis. I will also present a new Rh-group 13 catalyst system that is effective for the hydrodefluorination of challenging C-F bonds.