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Title: Electronically Unsaturated Three-Coordinate Aluminum Hydride and Organoaluminum Cations

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 $\textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate and electronically unsaturated aluminum hydride [LAIH]+[HB(C6F5)3]-(LH=1)]} \\ \textbf{Abstract:} \qquad \textbf{New three-coordinate aluminum hydride aluminum hyd$

[{(2,6-iPr2C6H3N)P(Ph2)}2N]H) and aluminum methyl [LAIMe]+[MeB(C6F5)3]— cations have been prepared. The quantitative estimation of Lewis acidity by Gutmann–Beckett method revealed [LAIH]+[HB(C6F5)3]— to be better Lewis acid than B(C6F5)3 and AlCl3 making these compounds ideal catalysts for Lewis acid-mediated reactions. To highlight that the work is of fundamental importance, catalytic hydroboration of aliphatic and aromatic aldehydes and ketones have been demonstrated. Important steps of the catalytic cycle have been probed by using multinuclear NMR measurements, including successful characterization of the proposed aluminum benzyloxide cationic intermediate, [LAI-O-CH2Ph]+[HB(C6F5)3]—. The proposed catalytic cycle has been found to be consistent with experimental observations and computational studies clearly indicating the migration of hydride from cationic aluminum center to the carbonyl carbon is the rate-limiting step

of the catalytic cycle.

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