Electronic Supplementary Information for:

# Subvalent Group 13 Molecules by Carbene-induced Hydrogen Abstraction

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**NMR spectra of compounds**

($Me_2Im^{Me}$)·Cp*$AlH_2$ **5**

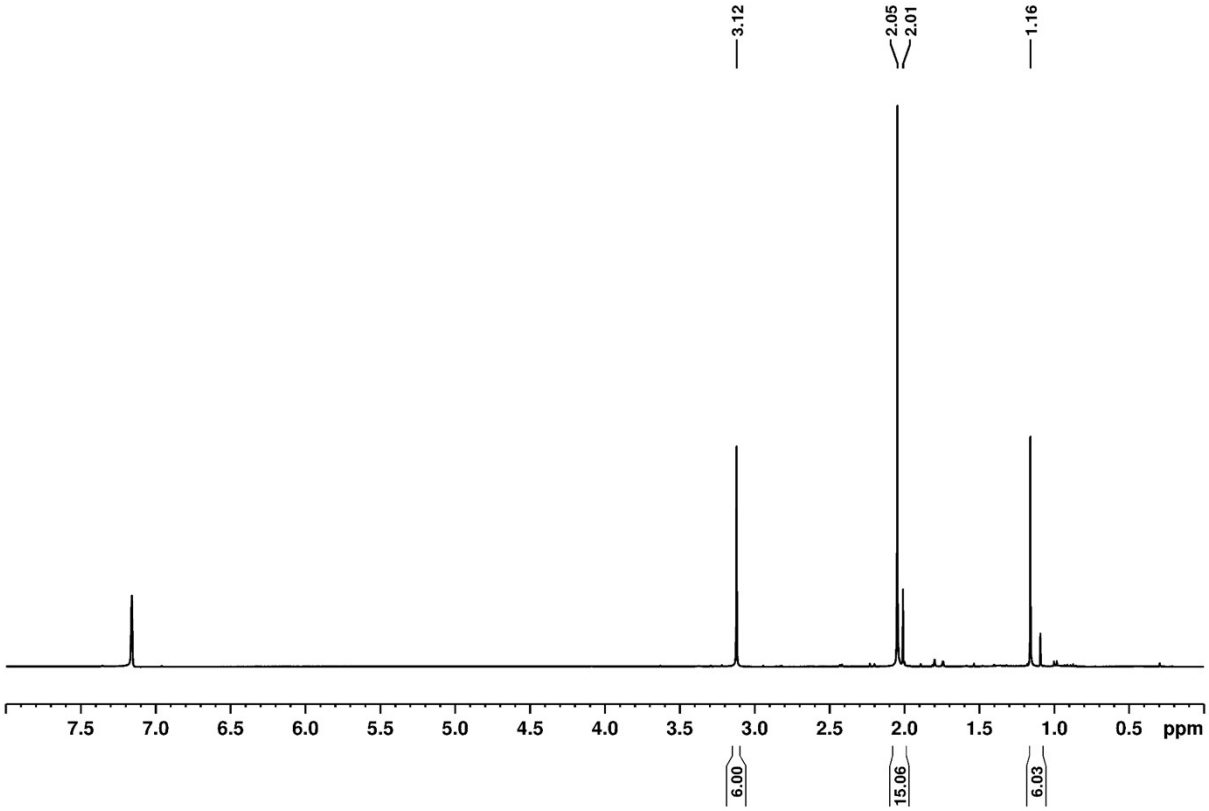


Figure S1. $^{1}H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·Cp*$AlH_2$ **5**

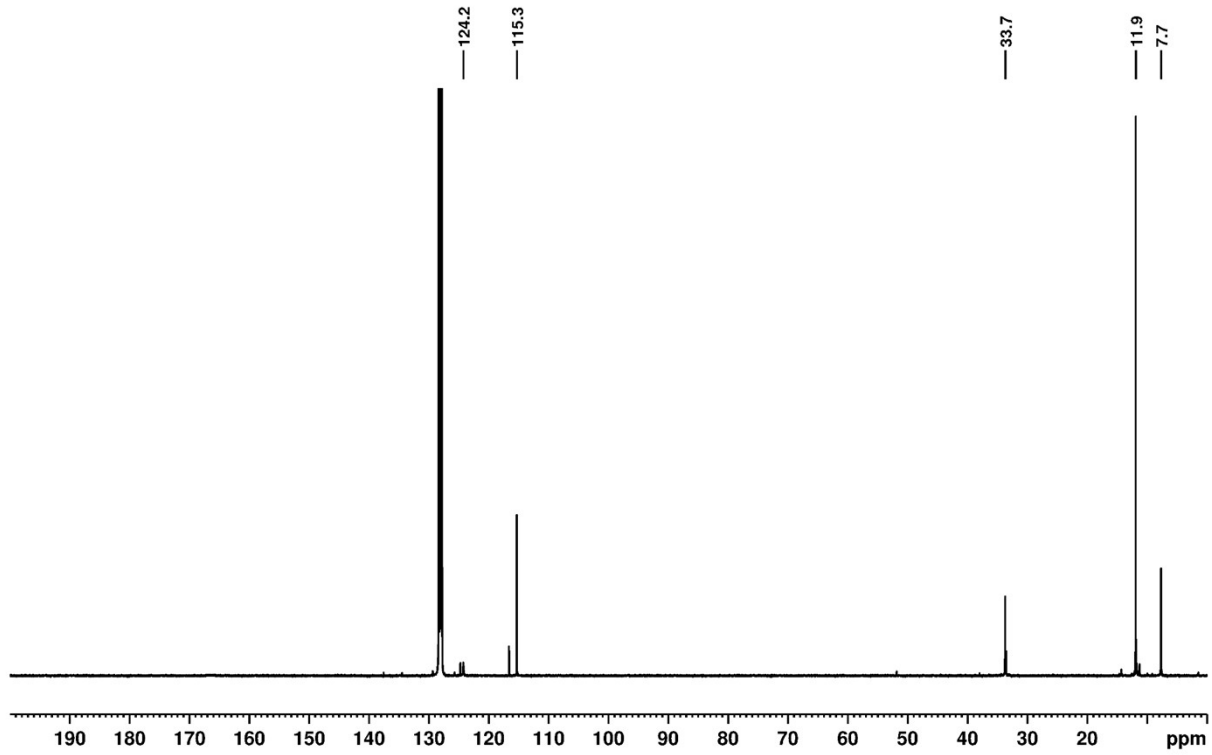


Figure S2. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·Cp*$AlH_2$ **5**.

($i$Pr$_2$Im$^{Me}$)·Cp*AlH$_2$ **6**:

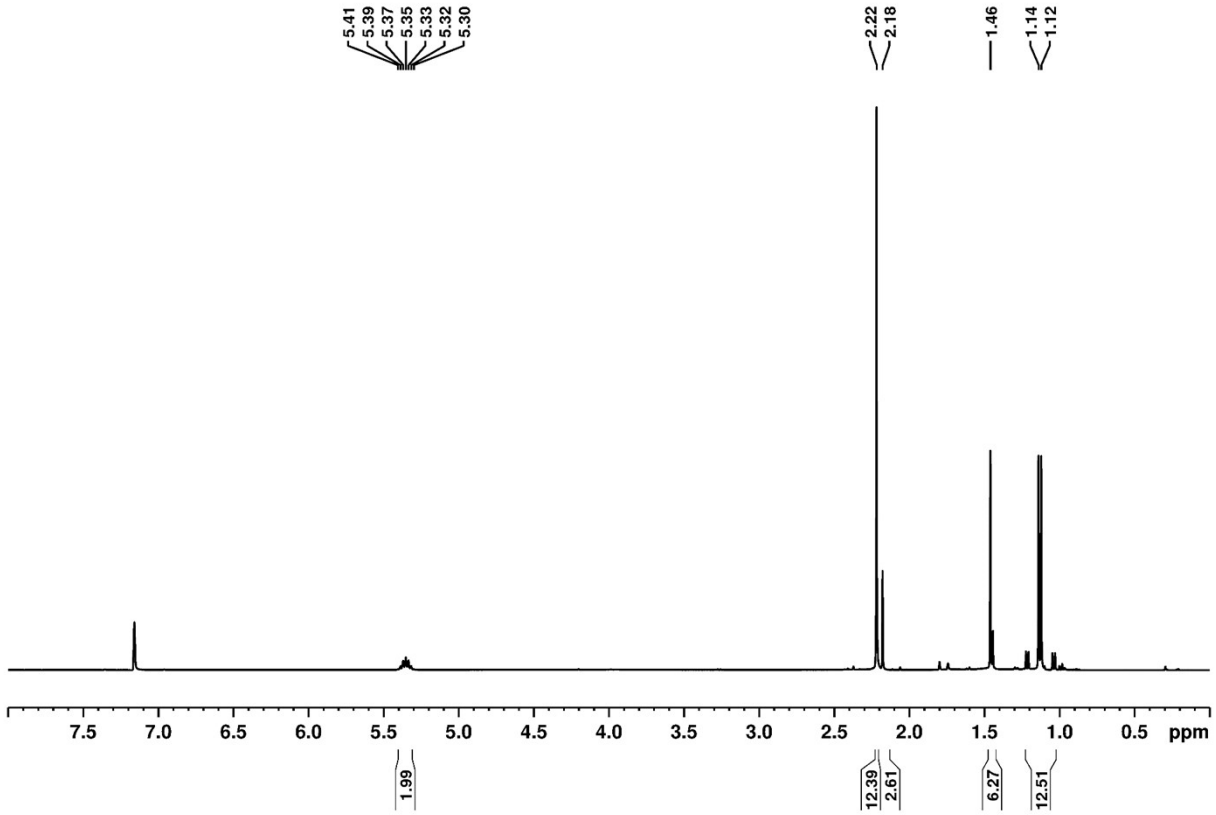


Figure S3. $^1$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($i$Pr$_2$Im$^{Me}$)·Cp*AlH$_2$ **6**.

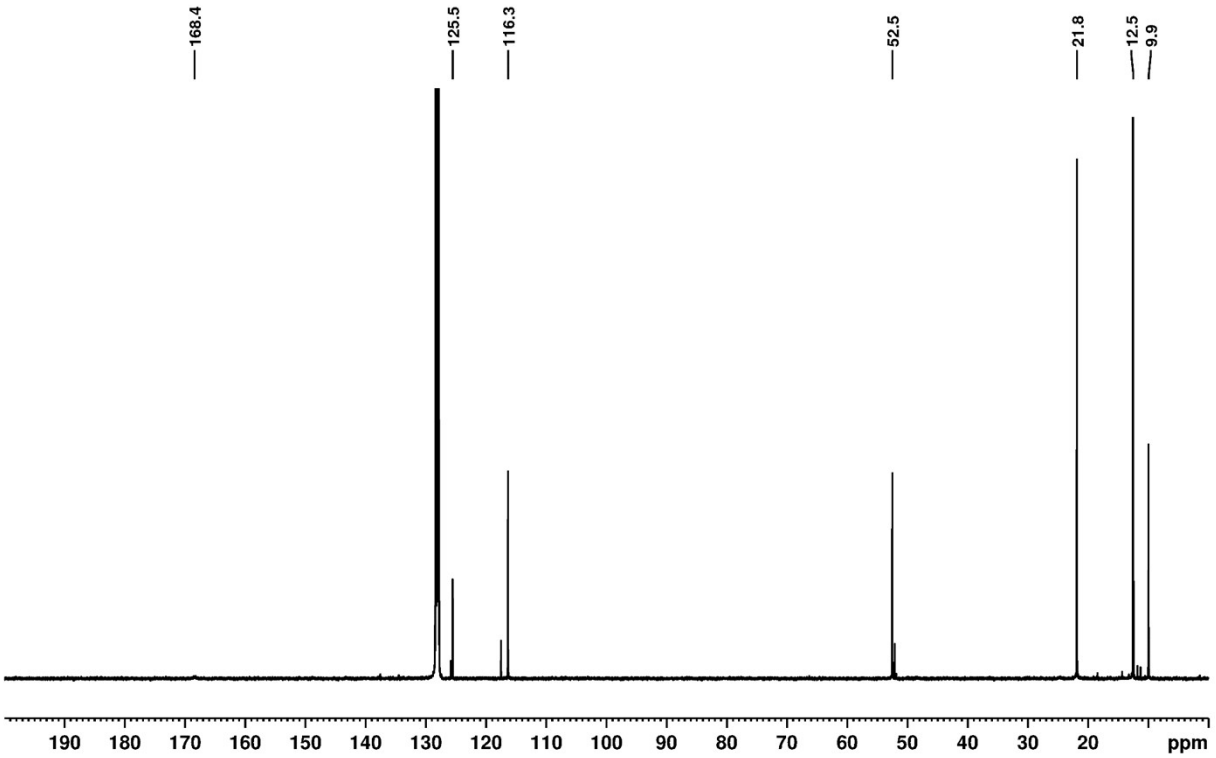


Figure S4. $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($i$Pr$_2$Im$^{Me}$)·Cp*AlH$_2$ **6**.

($Dipp_2Im$)·$Cp^*AlH_2$ **7**:

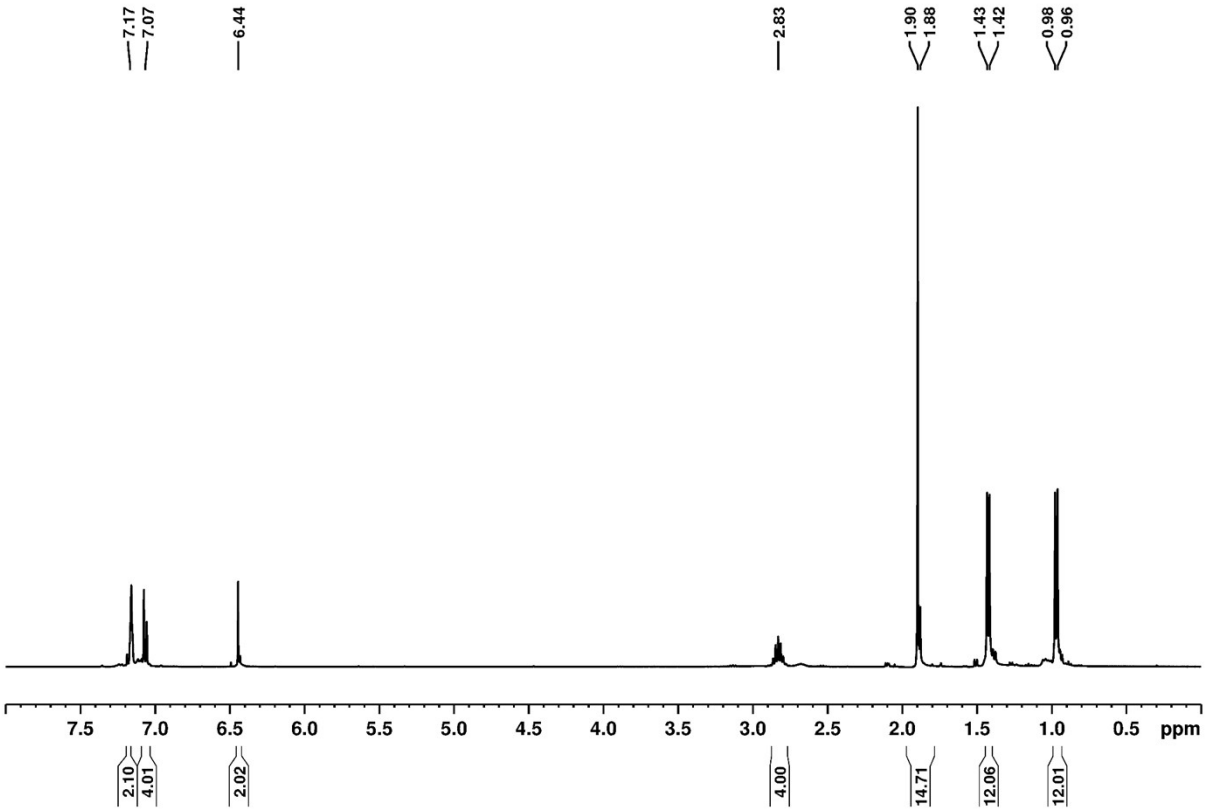


Figure S5. $^1H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($Dipp_2Im$)·$Cp^*AlH_2$ **7**.

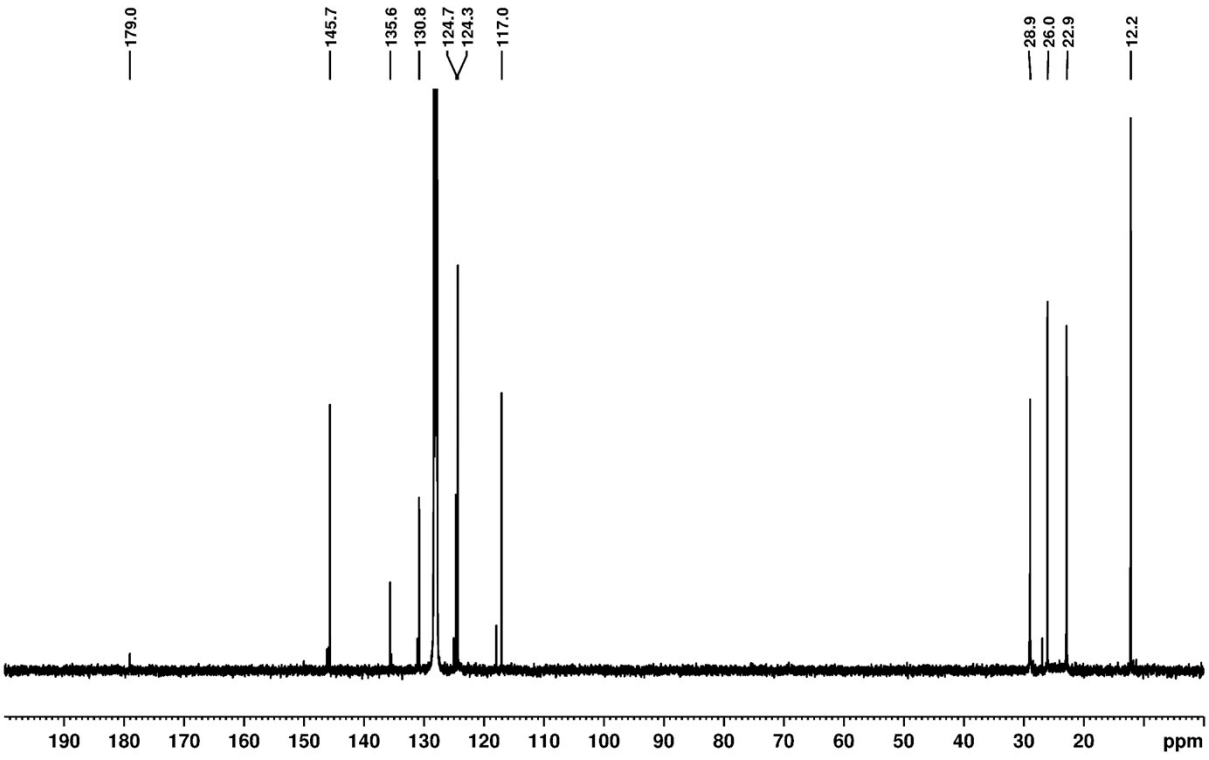


Figure S6. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($Dipp_2Im$)·$Cp^*AlH_2$ **7**.

($Me_2Im^{Me}$)·Cp*$GaH_2$ **8**:

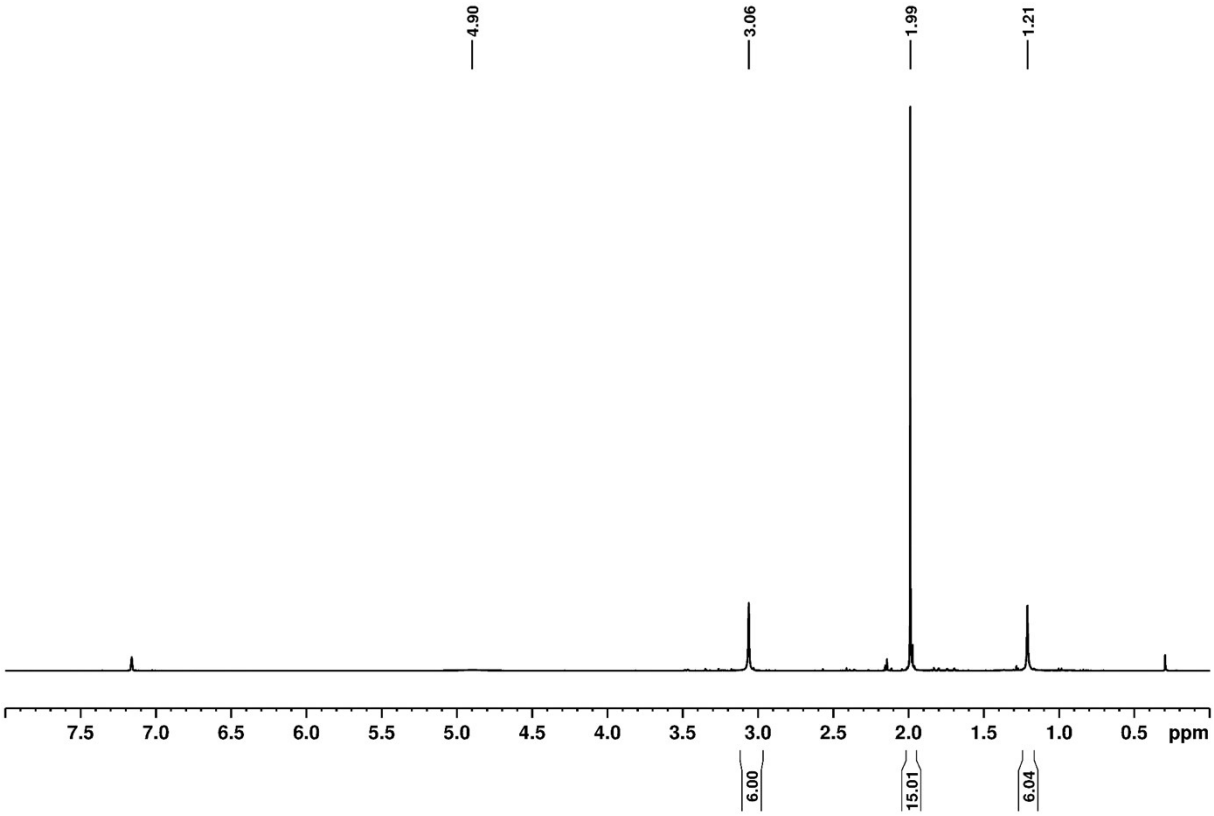


Figure S7. $^{1}H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·Cp*$GaH_2$ **8**.

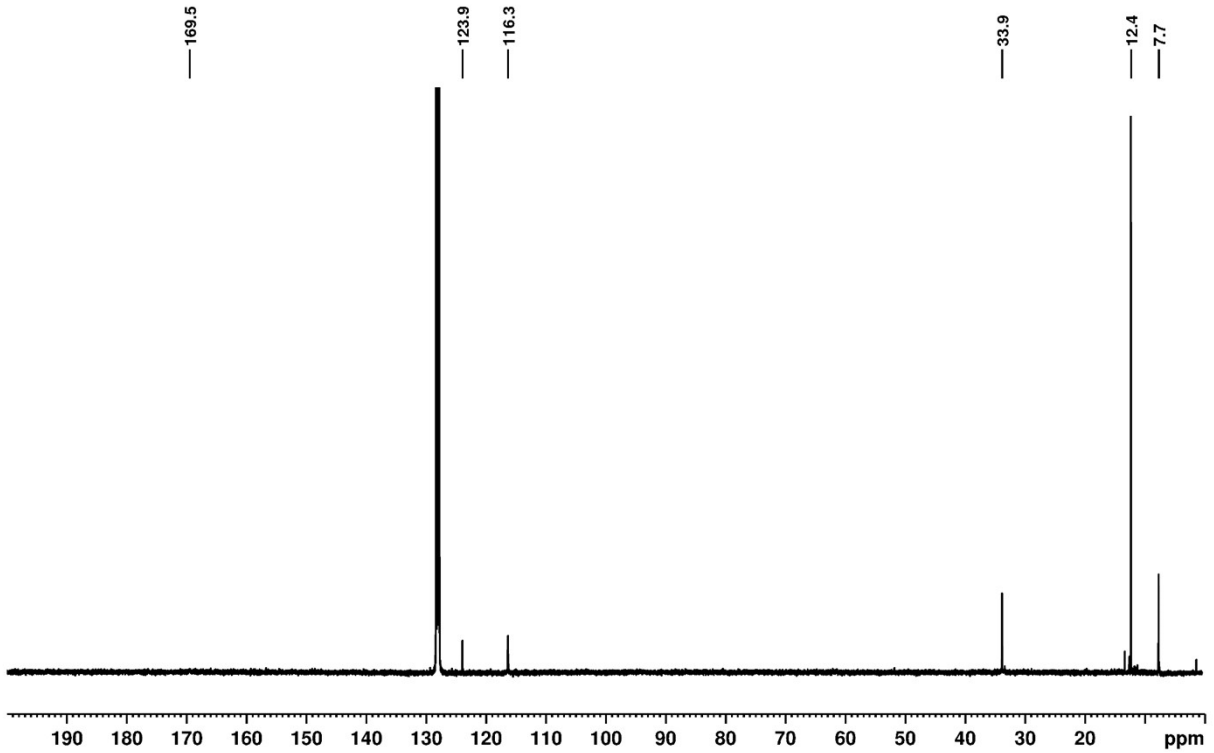


Figure S8. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·Cp*$GaH_2$ **8**.

(*i*Pr$_2$Im$^{Me}$)·Cp*GaH$_2$ **9**:

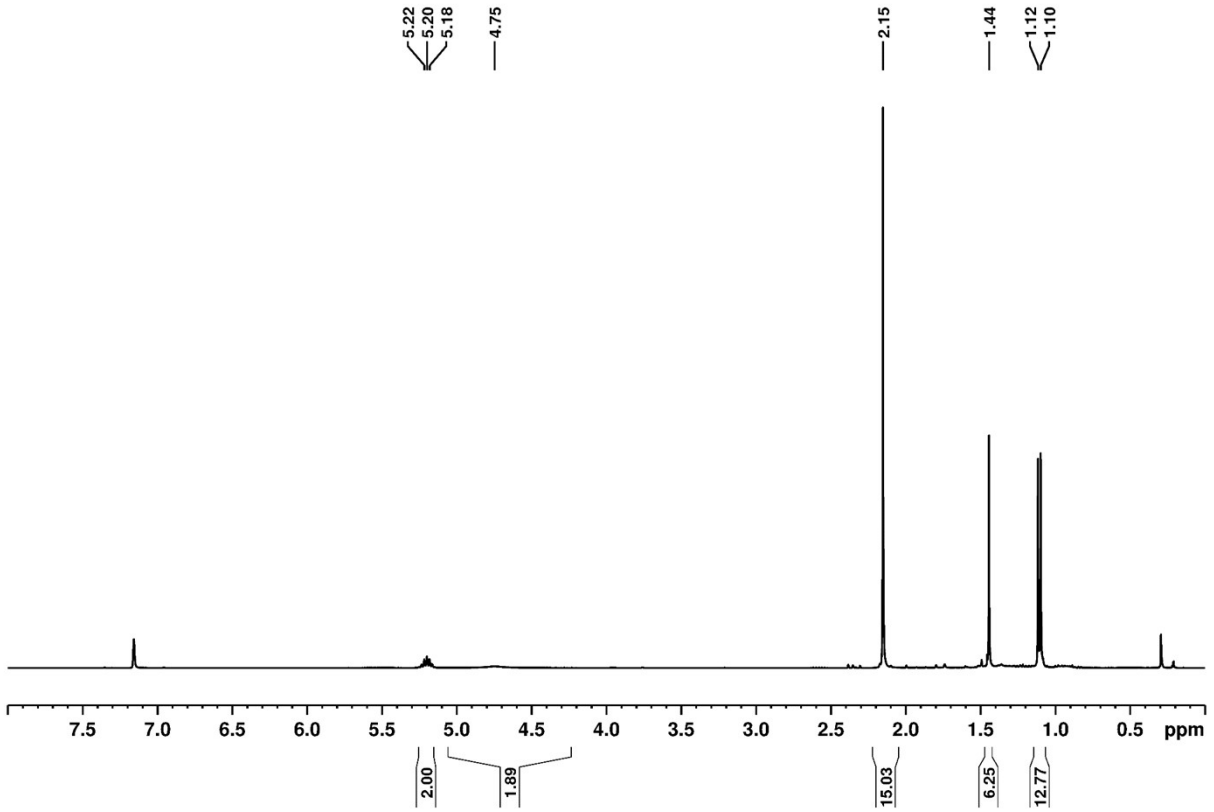


Figure S9. $^1$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of (*i*Pr$_2$Im$^{Me}$)·Cp*GaH$_2$ **9**.

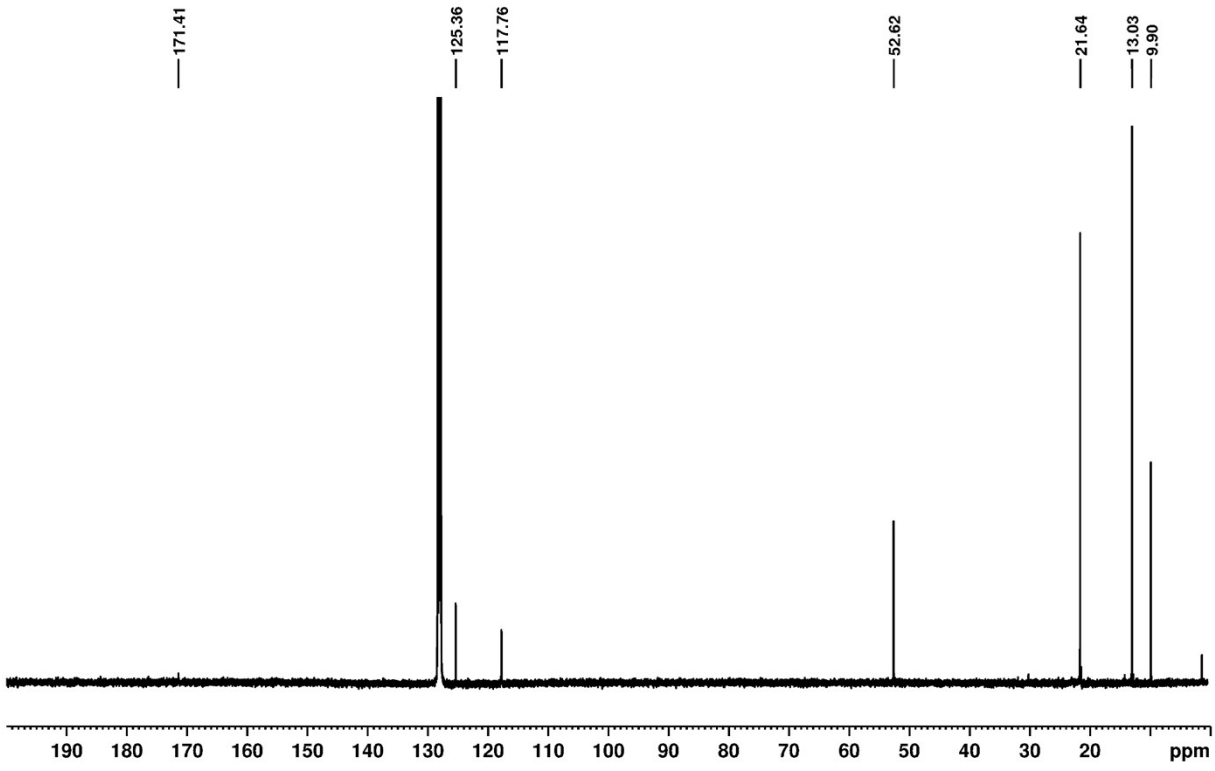


Figure S10. $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of (*i*Pr$_2$Im$^{Me}$)·Cp*GaH$_2$ **9**.

($Dipp_2Im$)·$Cp^*GaH_2$ **10**:

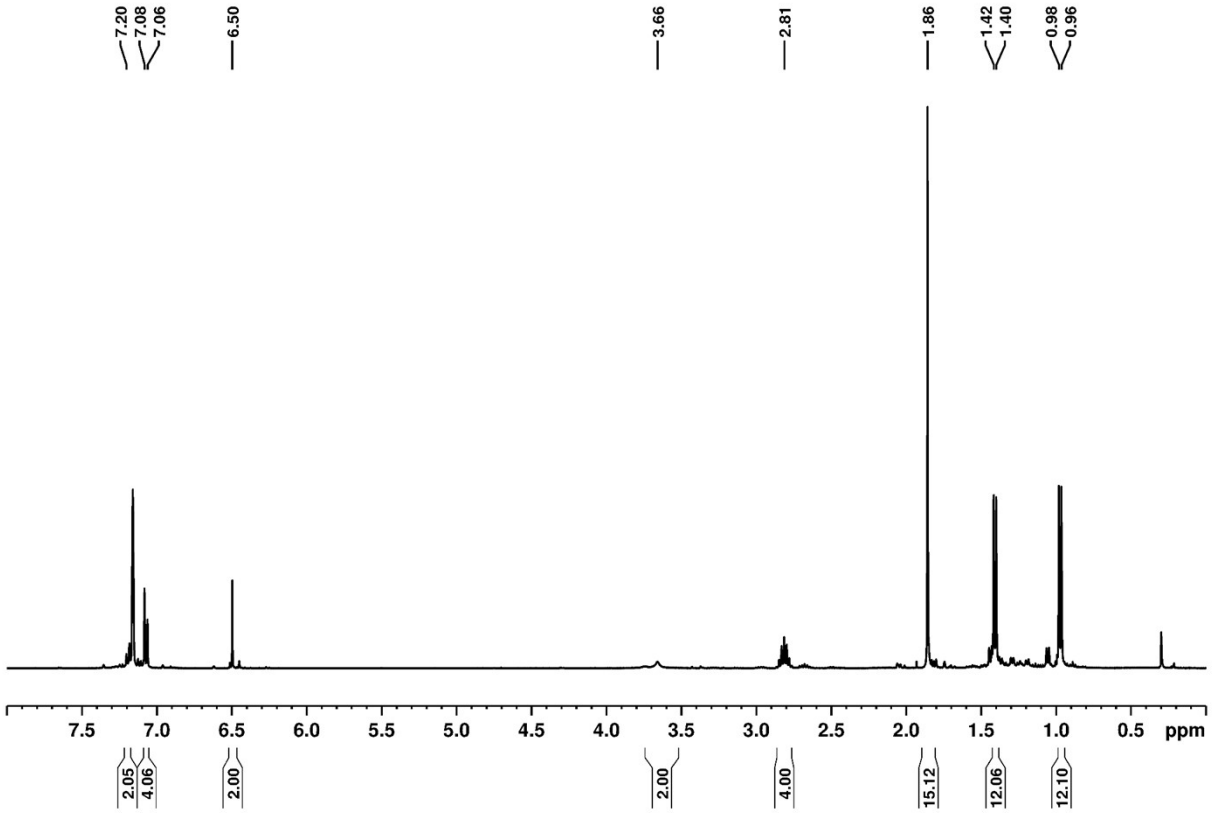


Figure S11. $^1H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($Dipp_2Im$)·$Cp^*GaH_2$ **10**.

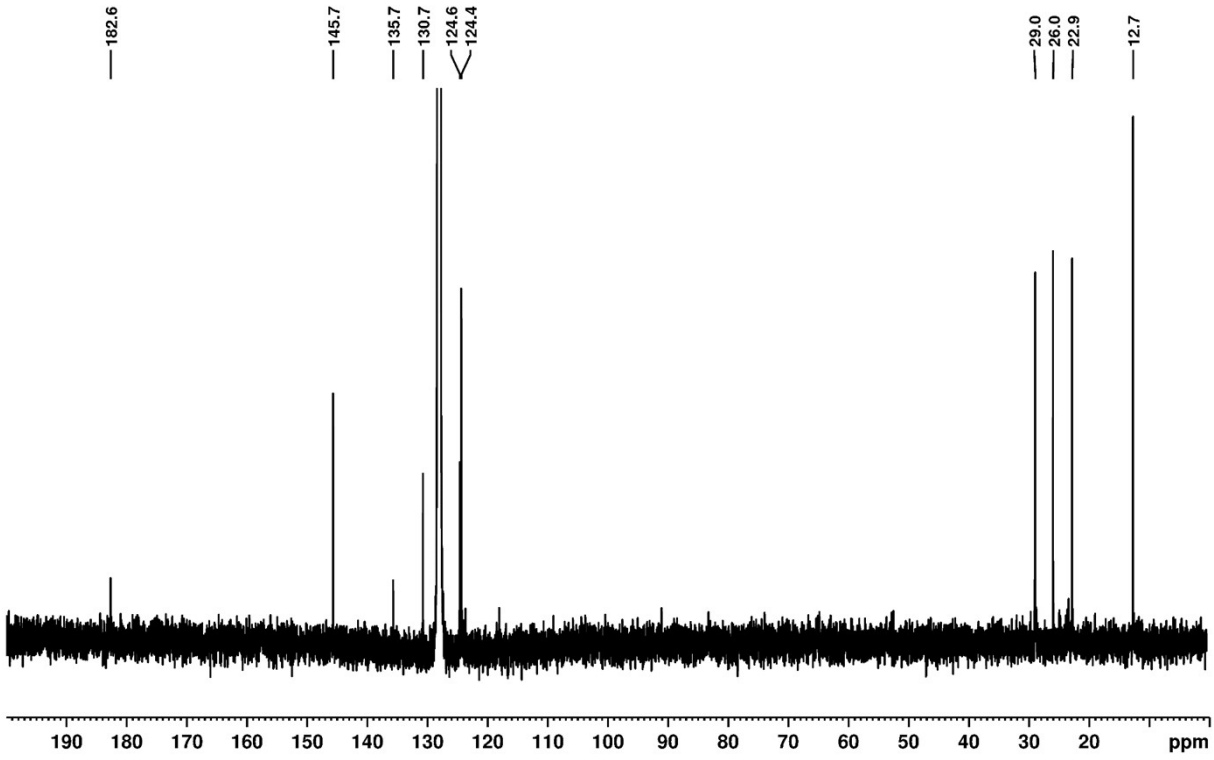


Figure S12. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($Dipp_2Im$)·$Cp^*GaH_2$ **10**.

(RER-Dipp$_2$Im$^H$H$_2$)·AlCp* **12**:

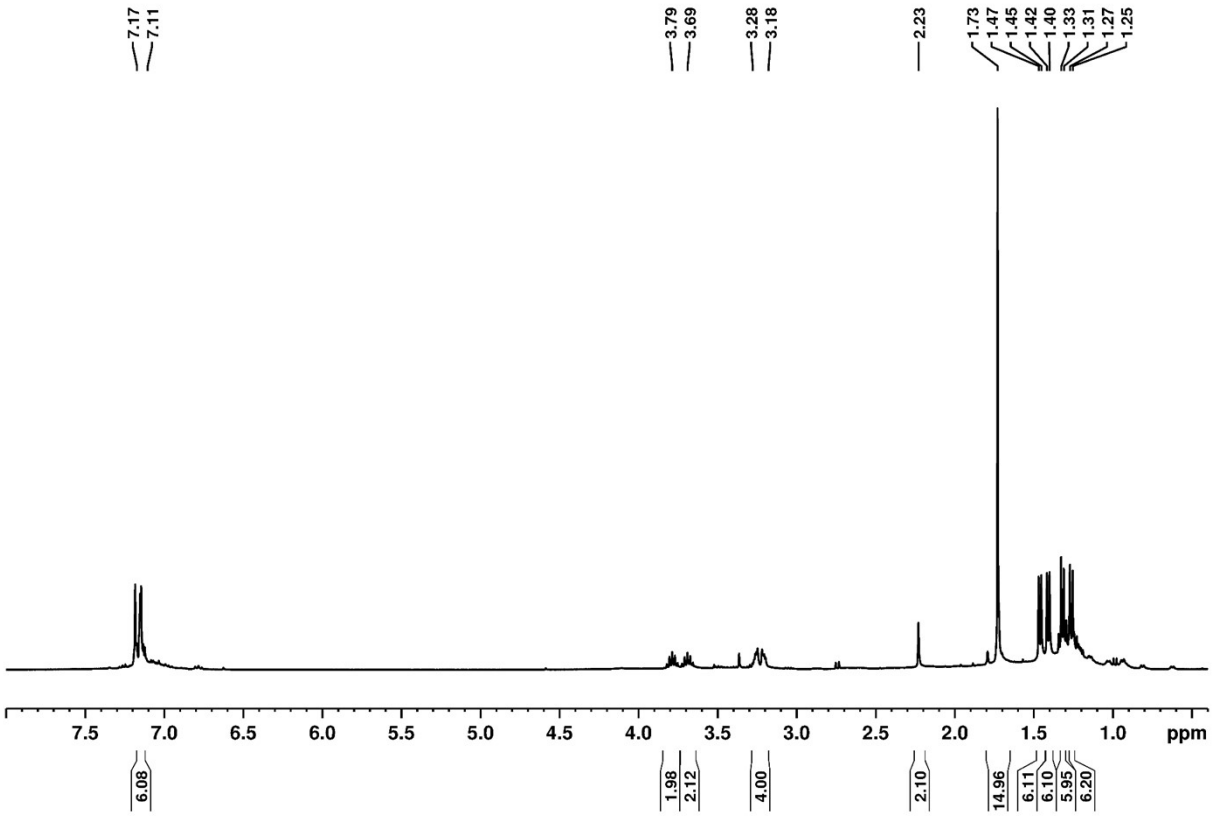


Figure S13. $^1$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of (RER-Dipp$_2$Im$^H$H$_2$)·AlCp* **12**.

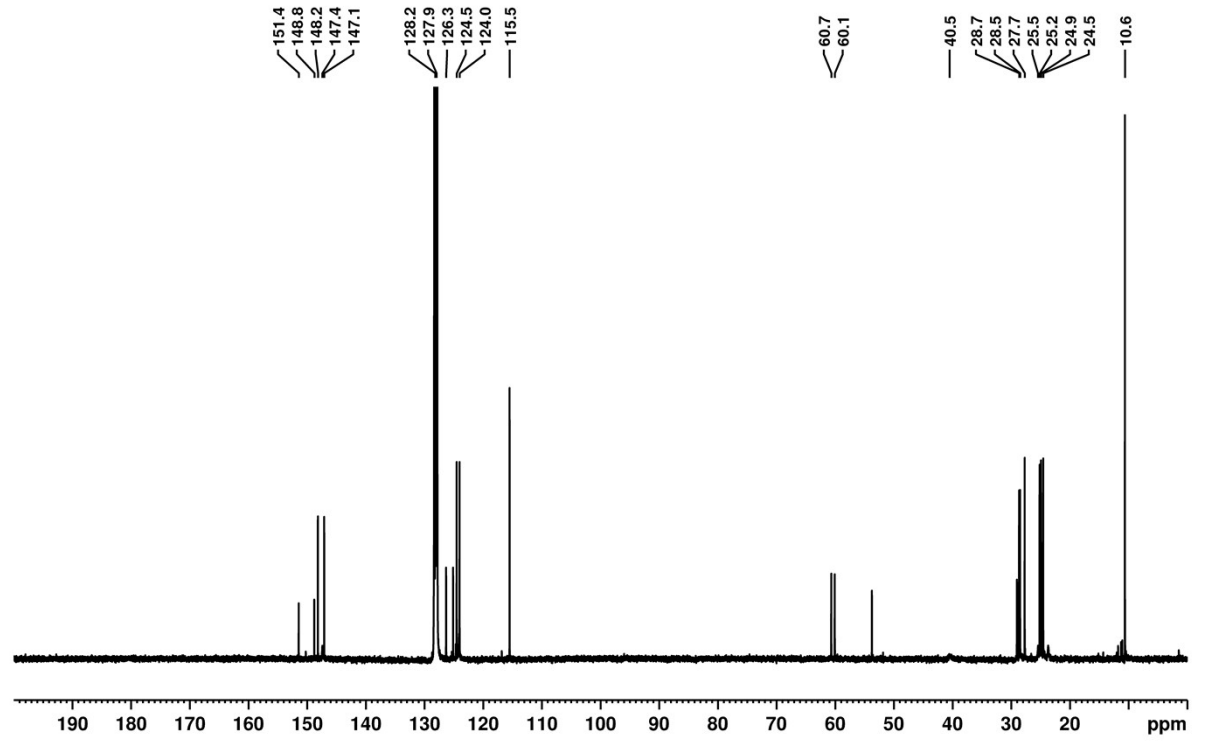


Figure S14: $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of (RER-Dipp$_2$Im$^H$H$_2$)·AlCp* **12**.

($Me_2Im^{Me}$)·AlCp*(RER-$Dipp_2Im^HH_2$) **13**:

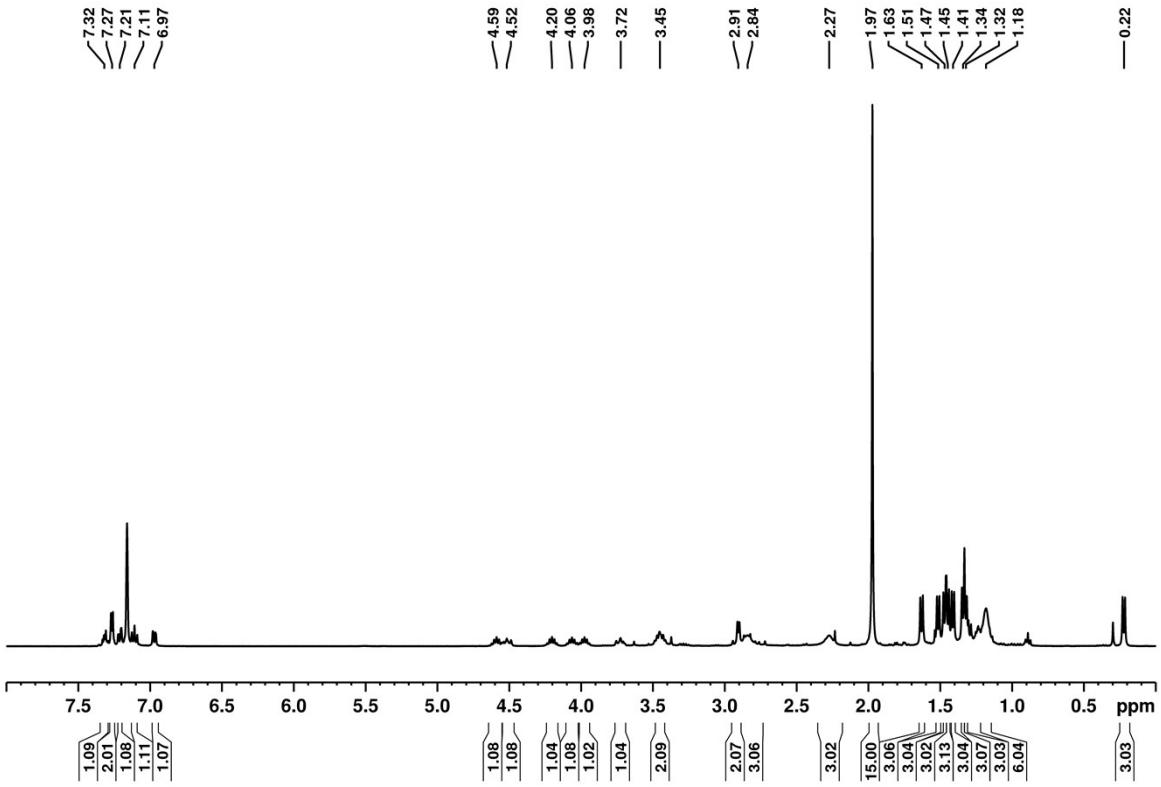


Figure S15. $^1$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·AlCp*(RER-$Dipp_2Im^HH_2$) **13**.

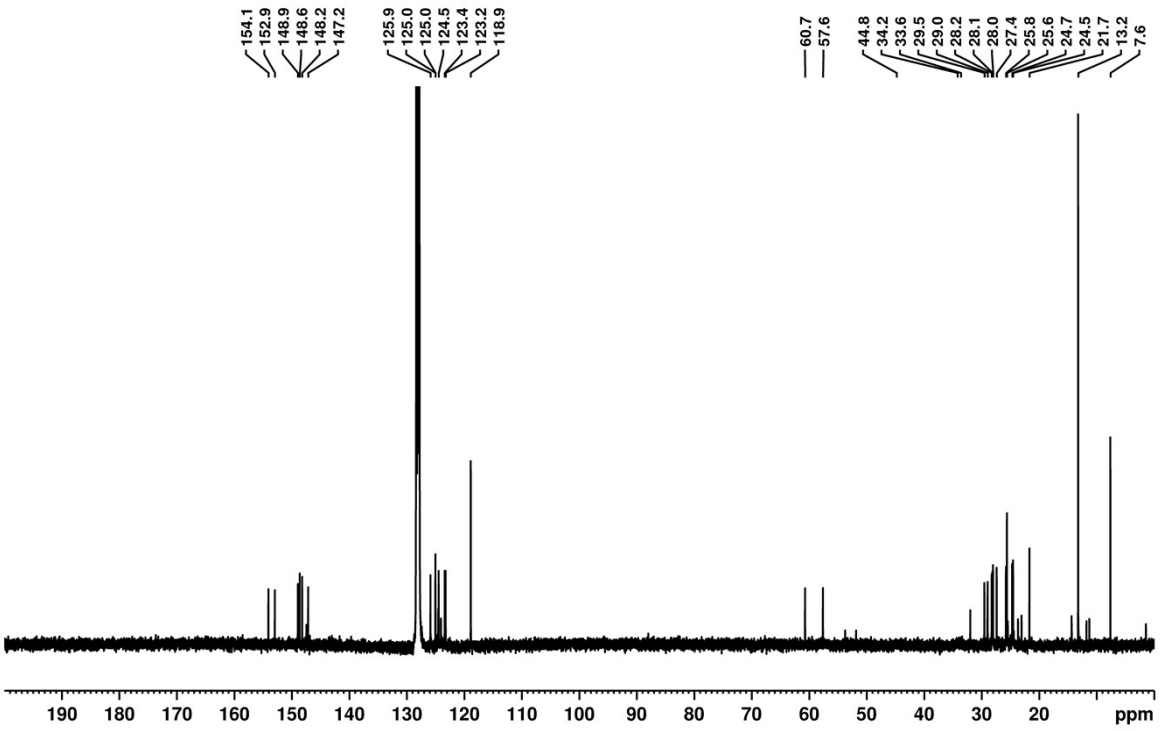


Figure S16. $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of ($Me_2Im^{Me}$)·AlCp*(RER-$Dipp_2Im^HH_2$) **13**.

*rac*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) ***rac*-14**:

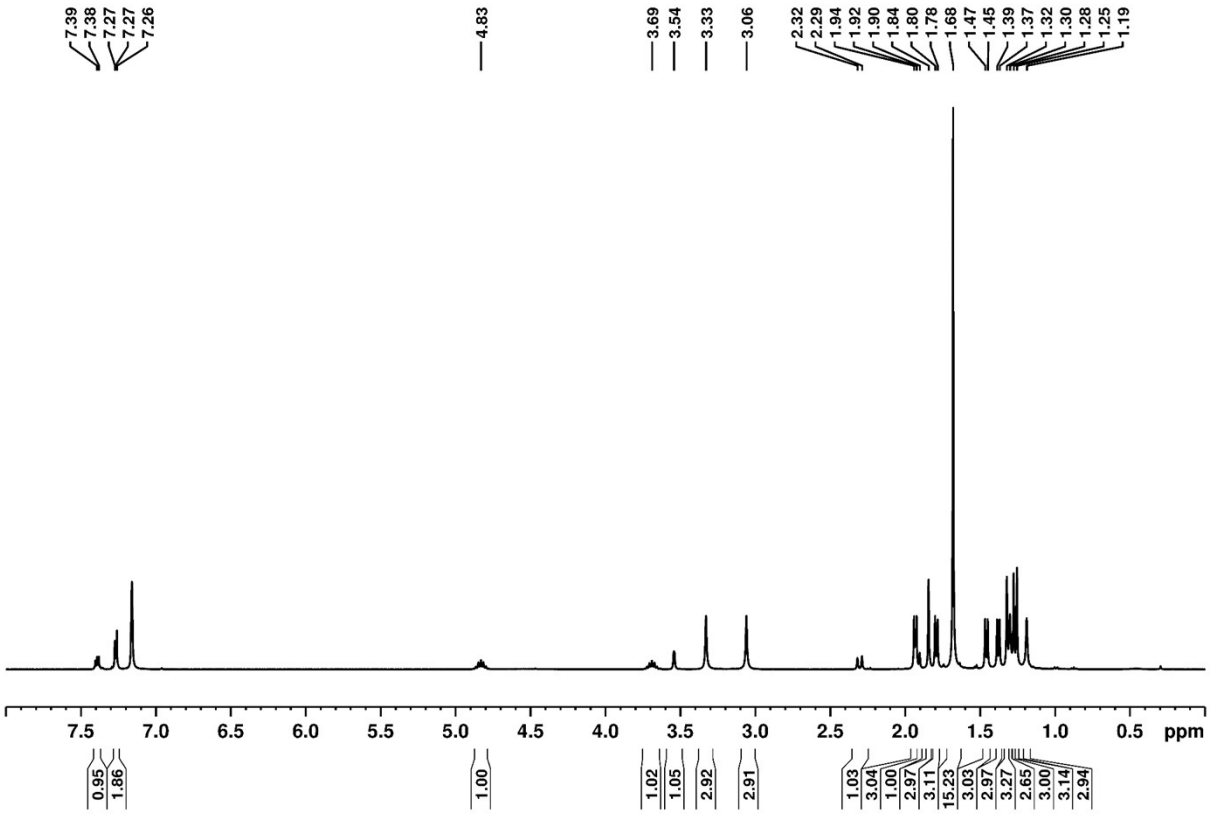


Figure S17. $^{1}H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of *rac*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) *rac*-**14**.

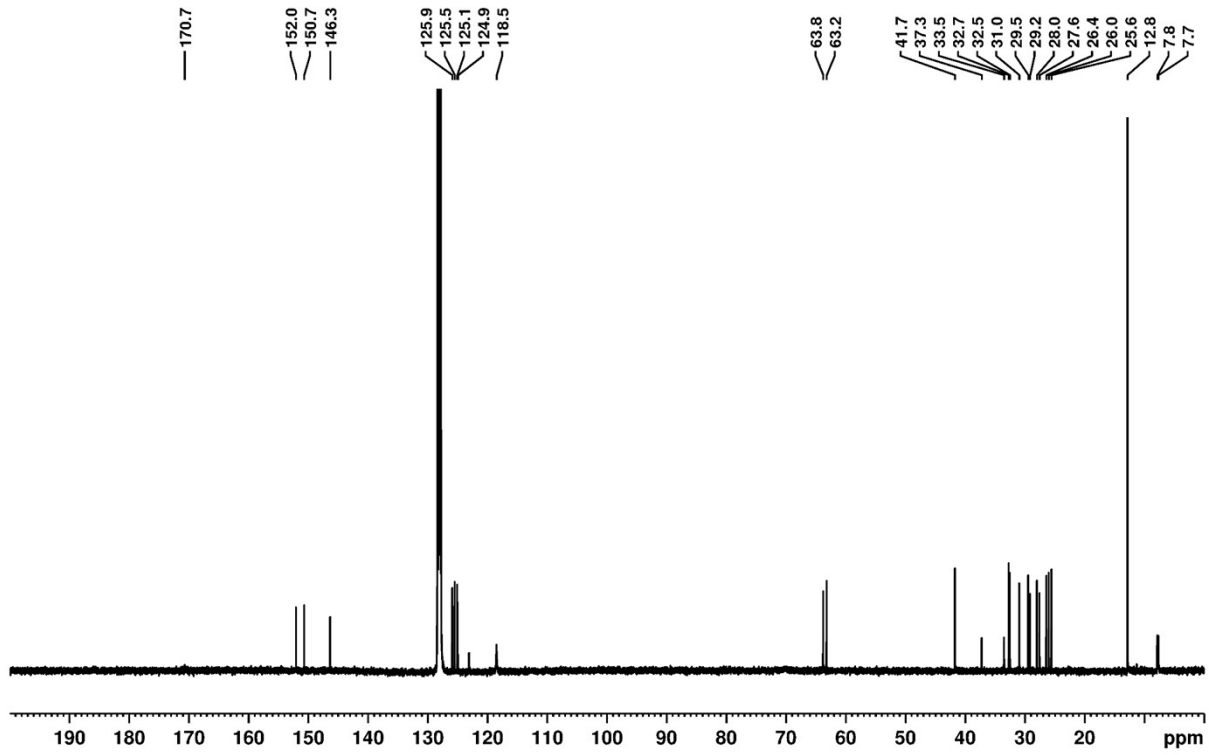


Figure S18. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of *rac*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) ***rac*-14**.

*meso*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) ***meso*-14**:

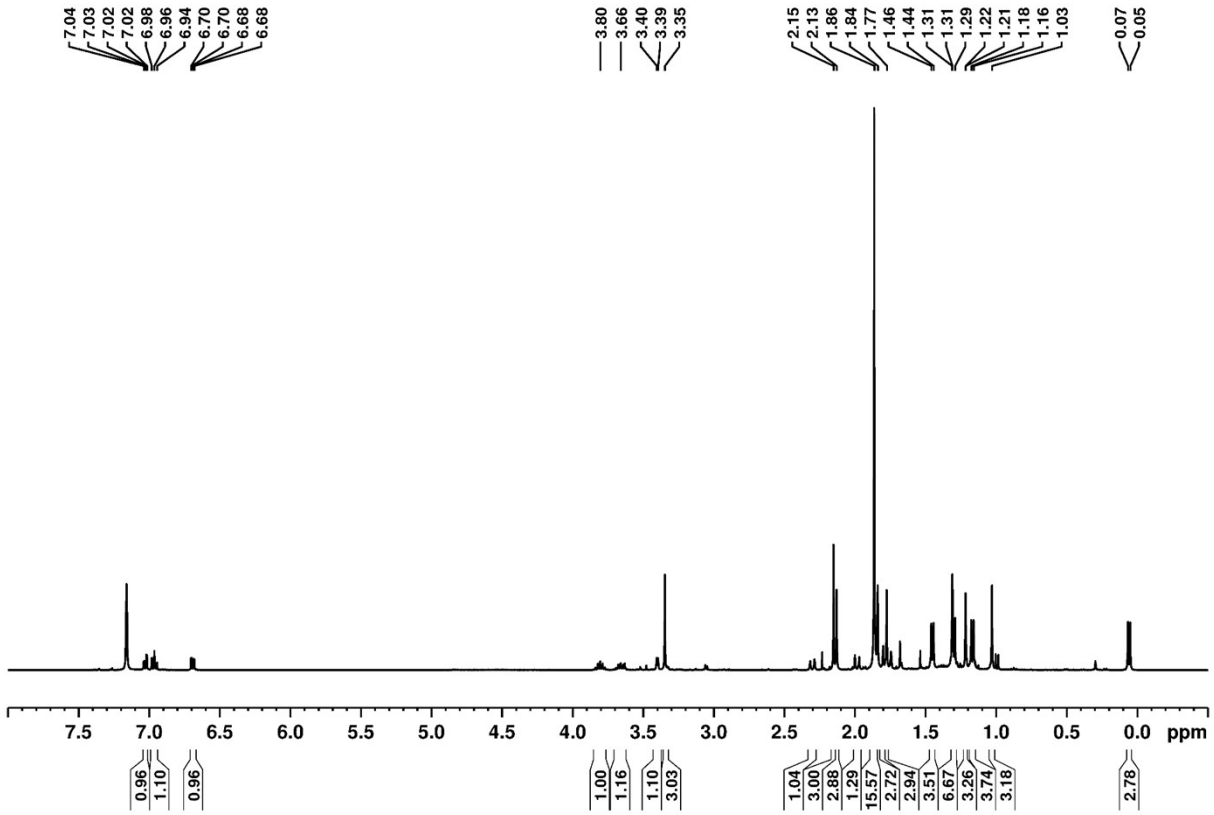


Figure S19. $^1$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of *meso*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) ***meso*-14**.

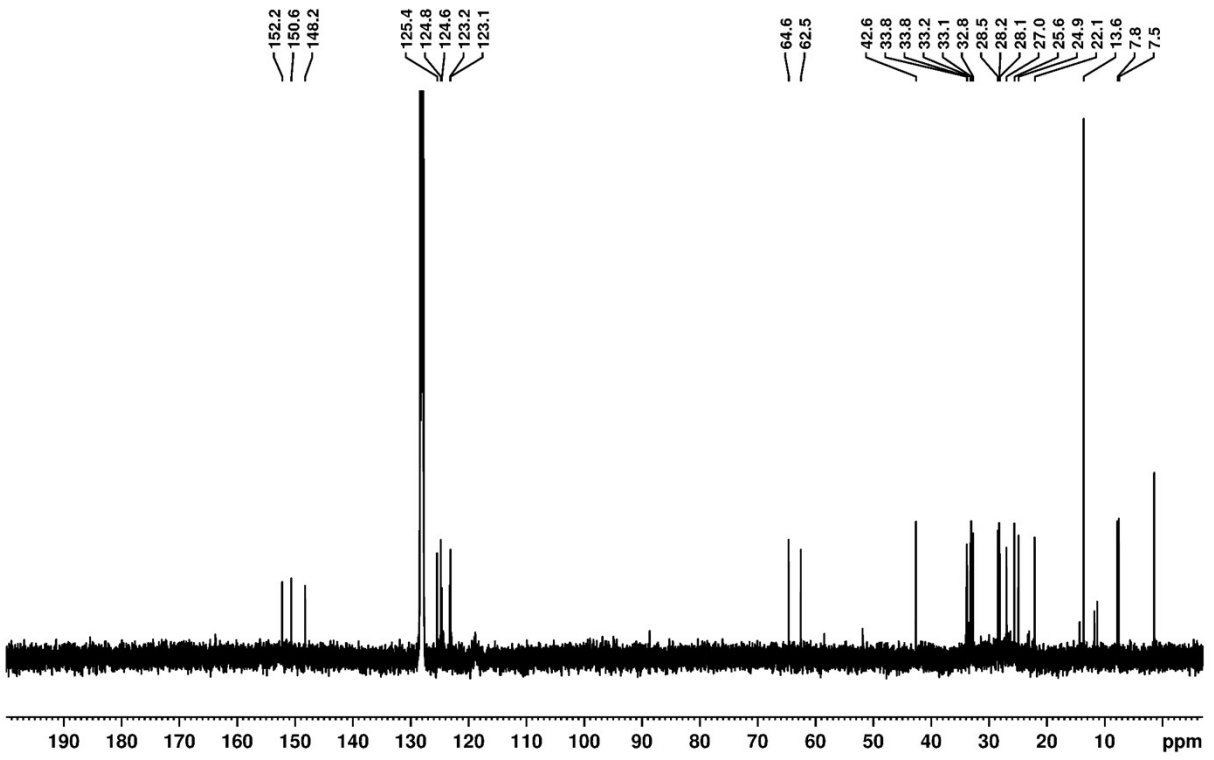


Figure S20. $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of *meso*-($Me_2Im^{Me}$)·AlHCp*($cAAC^{Me}H$) ***meso*-14**.

(cAAC$^{Me}$H)AlHCp* **15**:

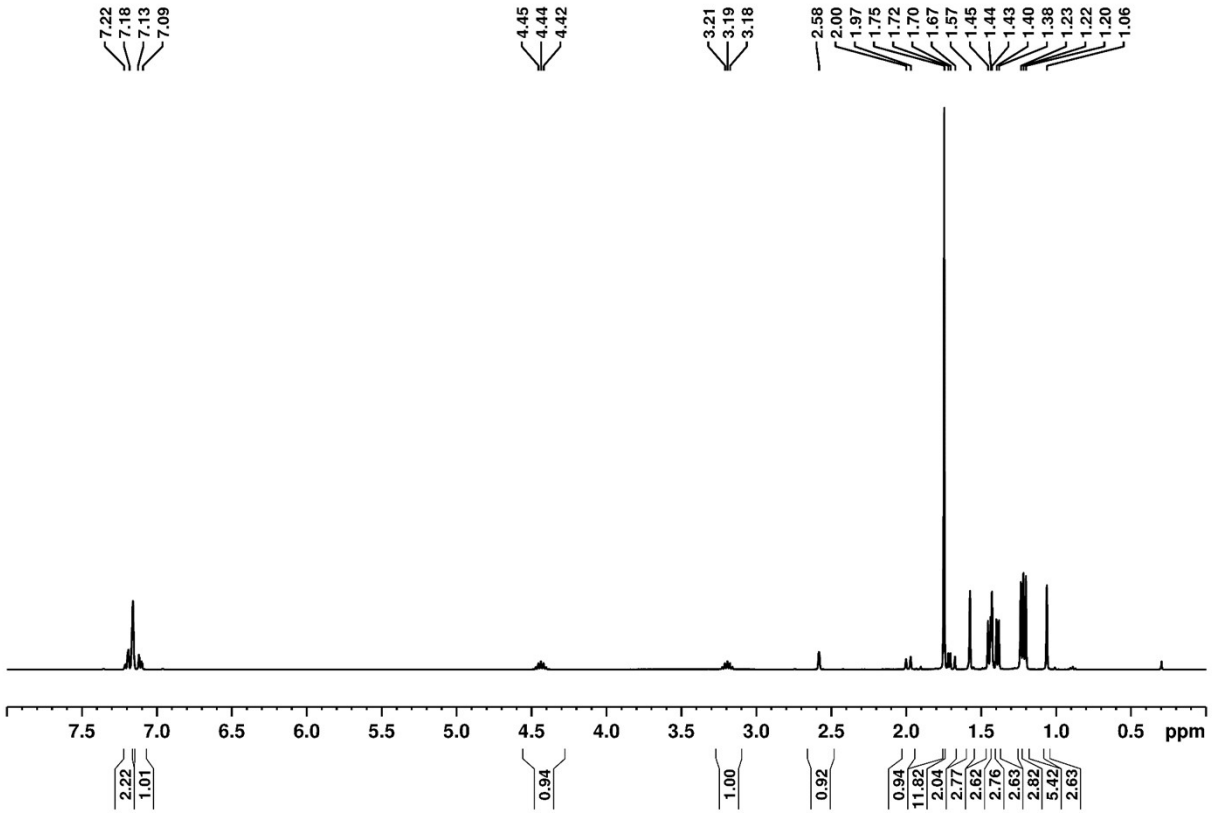


Figure S21. $^{1}$H NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of (cAAC$^{Me}$H)AlHCp* **15**.

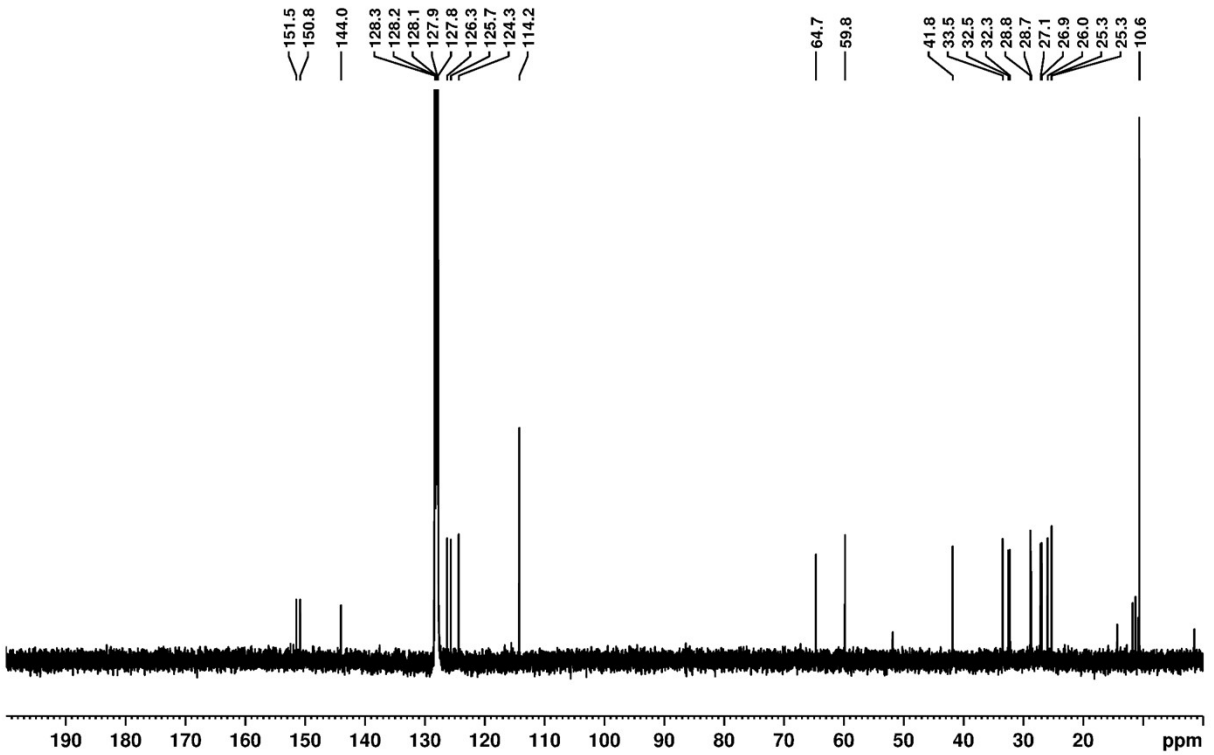


Figure S22. $^{13}$C NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of (cAAC$^{Me}$H)AlHCp* **15**.

Reaction of ($Dipp_2Im$)·$Cp^*GaH_2$ **10** with $cAAC^{Me}$:

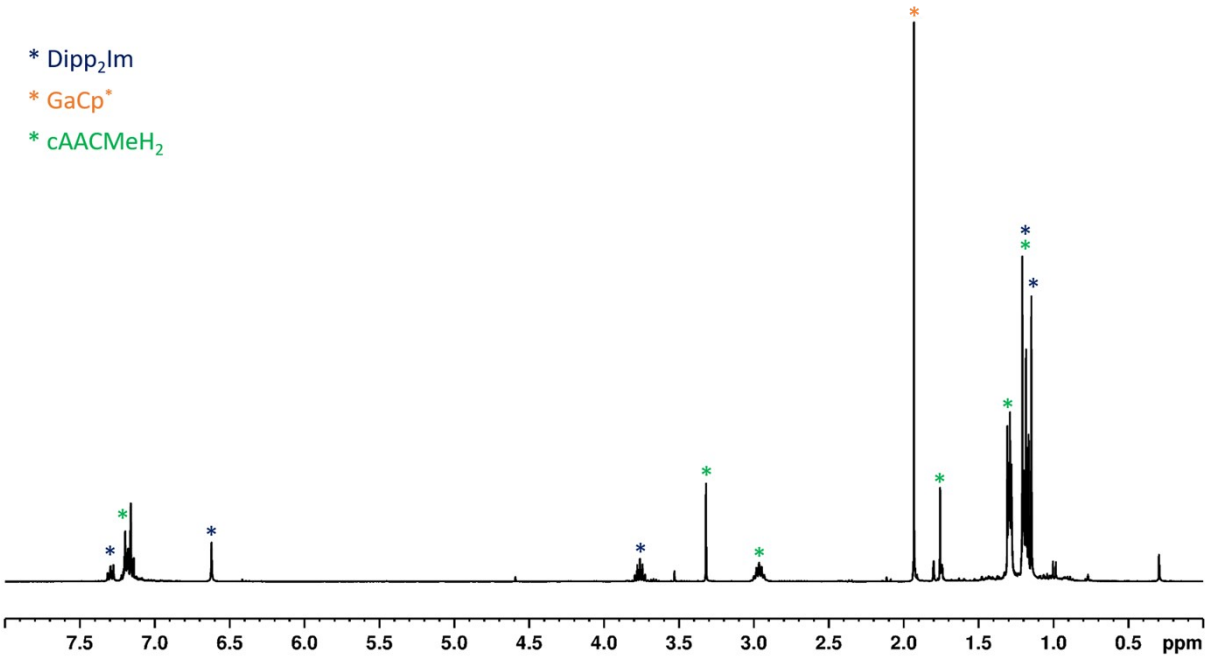


Figure S23. $^1H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of the reaction mixture of ($Dipp_2Im$)·$Cp^*GaH_2$ **10** with $cAAC^{Me}$ after 4 hours at 60°C. Signals marked with the blue asterisk belong to free $Dipp_2Im$, the signal with the orange asterisk to GaCp* and the signals with the green asterisk to $cAAC^{Me}H_2$.

GaCp* **16**:

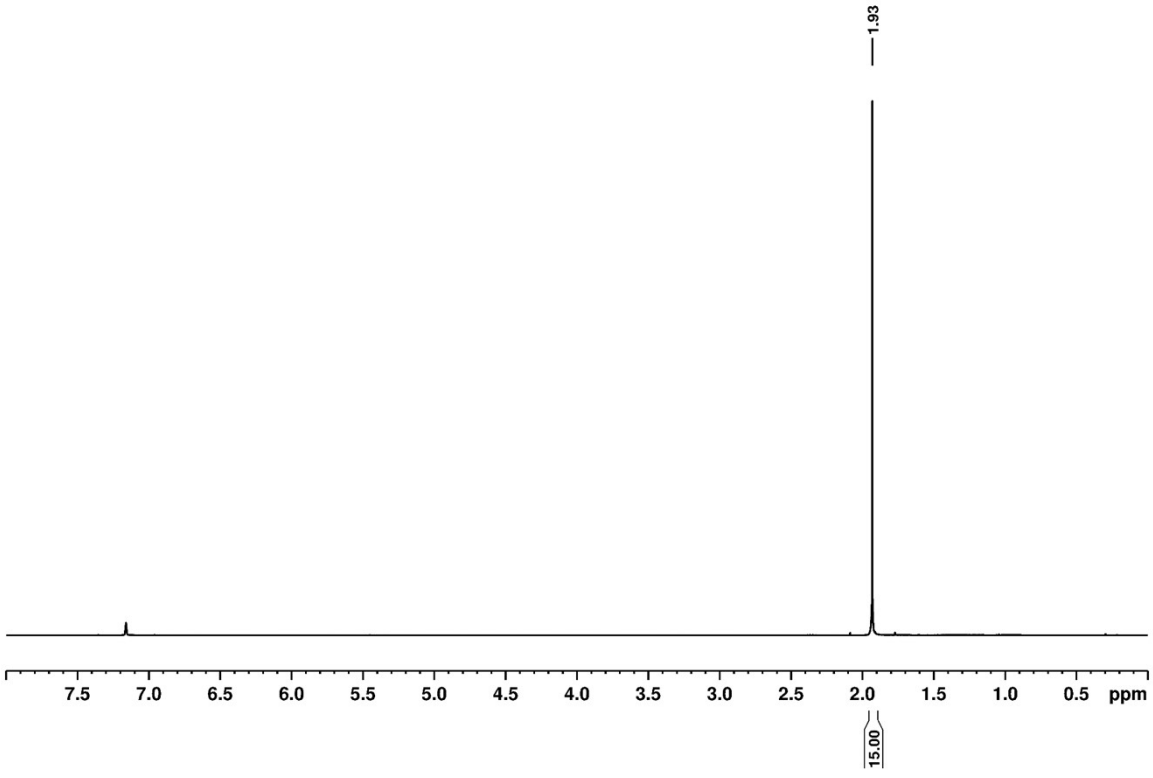

Figure S24. $^{1}H$ NMR spectrum (400.1 MHz, $C_6D_6$, 298 K) of GaCp* **16**.

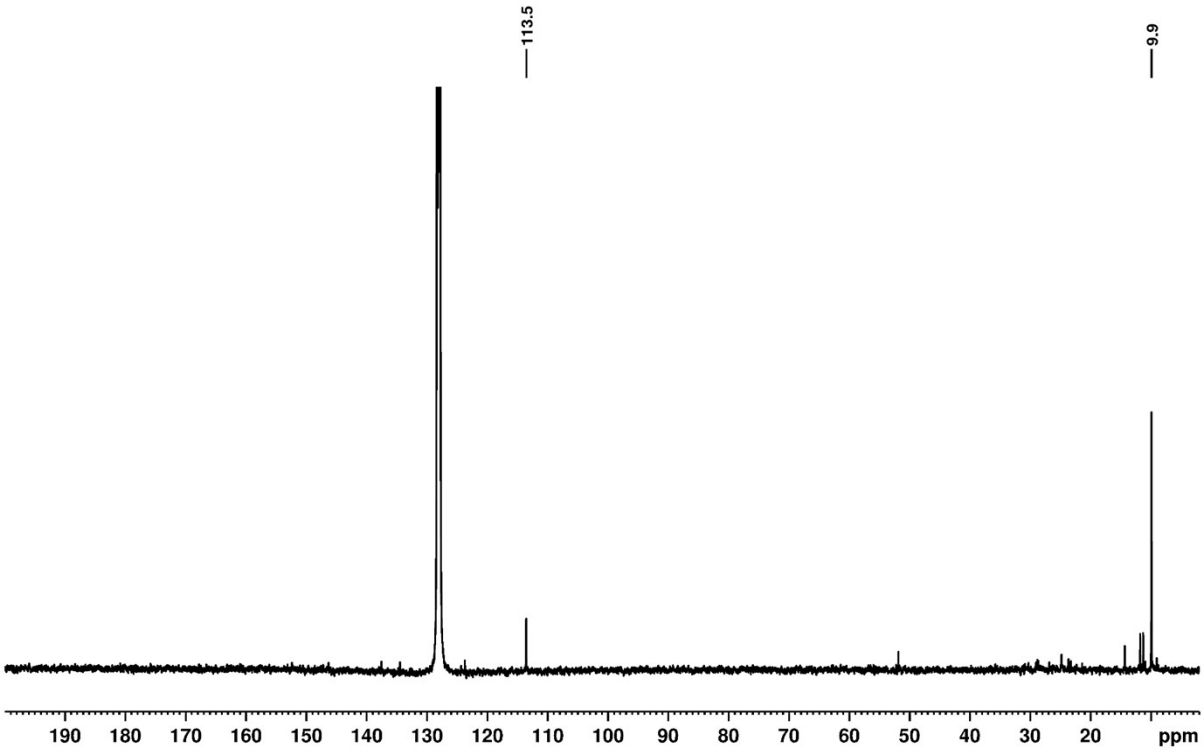


Figure S25. $^{13}C$ NMR spectrum (100.6 MHz, $C_6D_6$, 298 K) of GaCp* **16**.