The supplementary material mainly contains some additional graphical information from the main part in order to visualize the arguments given there. As in the main text, the naked metal atoms of the metalloid clusters are mainly presented in a metallic-grey colour and the surrounding shielding metal atoms are coloured depending on their formal oxidation state: blue  $(M^+)$ , orange  $(M^{2^+})$ .

The figures 1', 2', 3', 4' and 5' are presented on separate pages. The corresponding figure captions are:

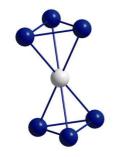
Fig. 1': The Al<sub>n</sub> core of the metalloid clusters  $[Al_7R_6]^-$ ,  $[Al_{12}R_6]^-$ ,  $[Al_{14}R_6I_6]^{2-}$  (R = N(SiMe<sub>3</sub>)<sub>2</sub>)

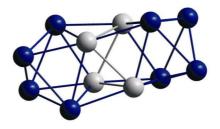
Fig. 2': The  $Al_n$  core of the metalloid clusters  $[Al_{69}R_{18}]^{3-}$  and  $[Al_{77}R_{20}]^{2-}$  ( $R = N(SiMe_2)_2$ ) and the different arrangements of the central  $Al_{13}$  units.

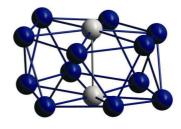
Fig. 3': The Al<sub>22</sub> core of the halides Al<sub>22</sub>X<sub>20</sub>·12D (X = Br, Cl; D = THF)

Fig. 4': Section of the normal-pressure  $(\alpha, \beta, \gamma, \delta)$  and the high-pressure modifications (GaII, GaIII) of solid Ga. The fcc high-pressure modification GaIV is not shown.

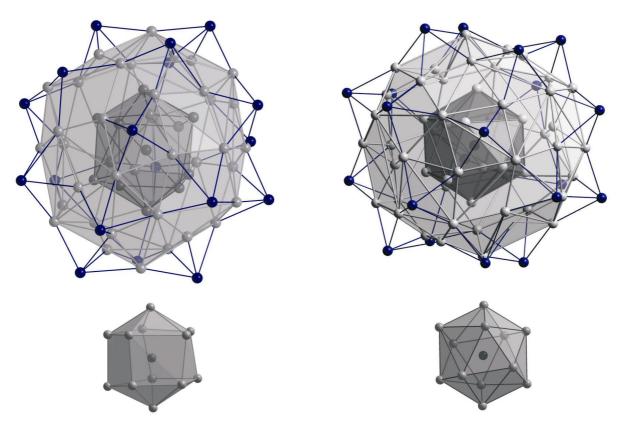
Fig. 5':  $Ga_n$  core of the clusters  $Ga_{18}R_8$  and  $Ga_{22}R_8$  ( $R = SitBu_3$ ): Furthermore the directly bonded Si atoms are shown (red). Within a similar cage of these surrounding atoms (8 Si or better the ligand-bearing 8 Ga) there are 10 resp. 14 Ga atoms arranged similar to these in normal-pressure  $\beta$ -Ga resp. high-pressure GaIII.



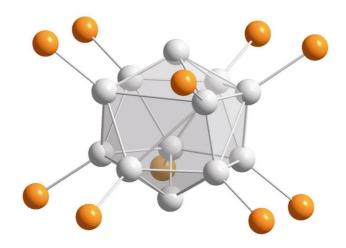




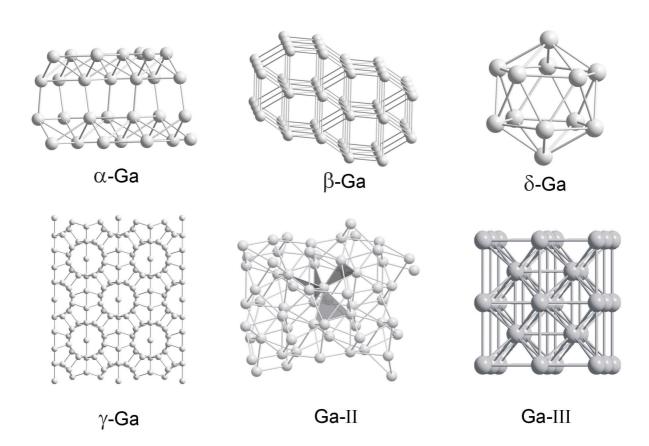
Supplement Figure 1



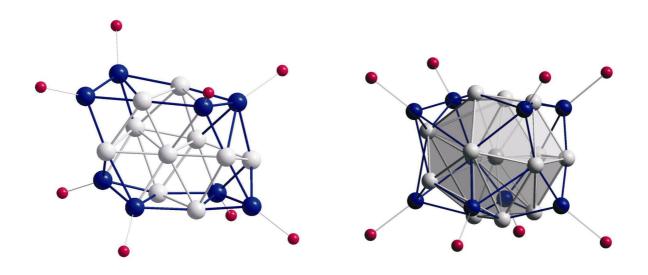
Supplement Figure 2



Supplement Figure 3



Supplement Figure 4



Supplement Figure 5