Electronic Supplementary Material (ESI) for Chemical Communications.

Supplementary Information: Thickness dependent thermal stability of 2D gallenene †

Krista G. Steenbergen*[a] and Nicola Gaston[b]

[a] The MacDiarmid Institute for Advanced Materials and Nanotechnology, School of Chemical and Physical Sciences, Victoria University of Wellington, P.O. Box 600, Wellington, New Zealand; E-mail: kgsteen@gmail.com
[b] The MacDiarmid Institute for Advanced Materials and Nanotechnology, Department of Physics, The University of Auckland, Private Bag 92019, Auckland, New Zealand.

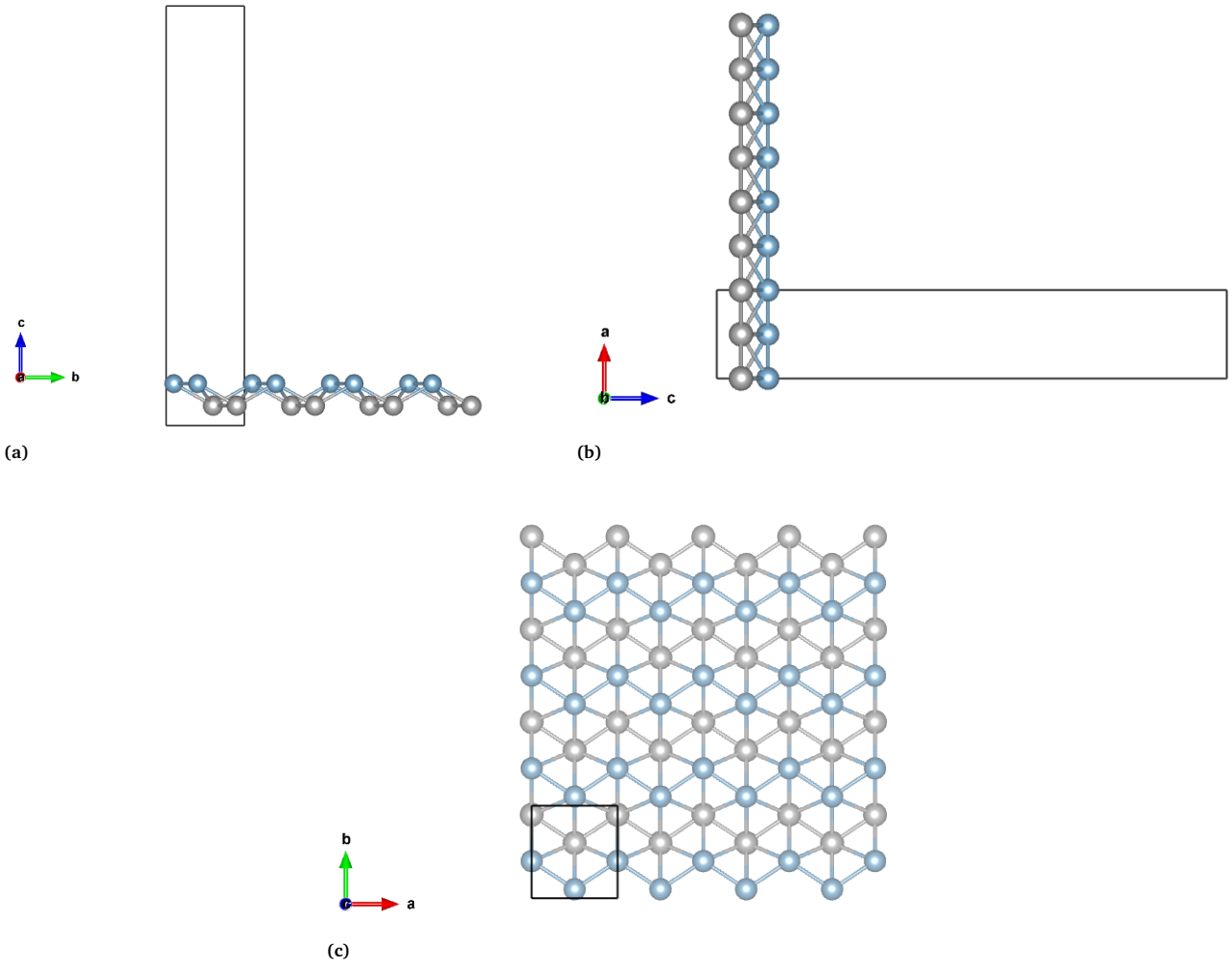
1 Optimised structure coordinates

Structural information is given in VASP POSCAR format.

```
  Optimised gallium trilayer
  1.00
     4.98032     0.00000     0.00000
     0.00000     2.84812     0.00000
    -1.35642    -1.08132    28.00000
 Ga
  6
Direct
     0.2537281   0.4547094   6.5790781
     0.3947621   1.8766707   4.1877088
     0.5358987   0.4547094   1.7962702
     0.7537265   1.8766707   6.5790781
     0.8947766   0.4547094   4.1877088
     0.0358971   1.8766707   1.7962702


  Optimised gallium bilayer
  1.00
     4.413321    0.000000    0.000000
     0.000000    4.763338    0.000000
     0.000000    0.000000   25.000000
 Ga
  4
Direct
     0.00000000  4.29509020  1.21436334
     0.00000000  1.91377974  2.55396413
     2.20666099  2.84962434  1.21436334
     2.20666099  0.46851102  2.55396413
```

2 Structural analysis



(a) (b) (c)

Fig. S1 Visualisations of the optimised bilayer structure aligned to unit cell axis. The bottom layer is shown in grey, with the upper layer shown in blue.

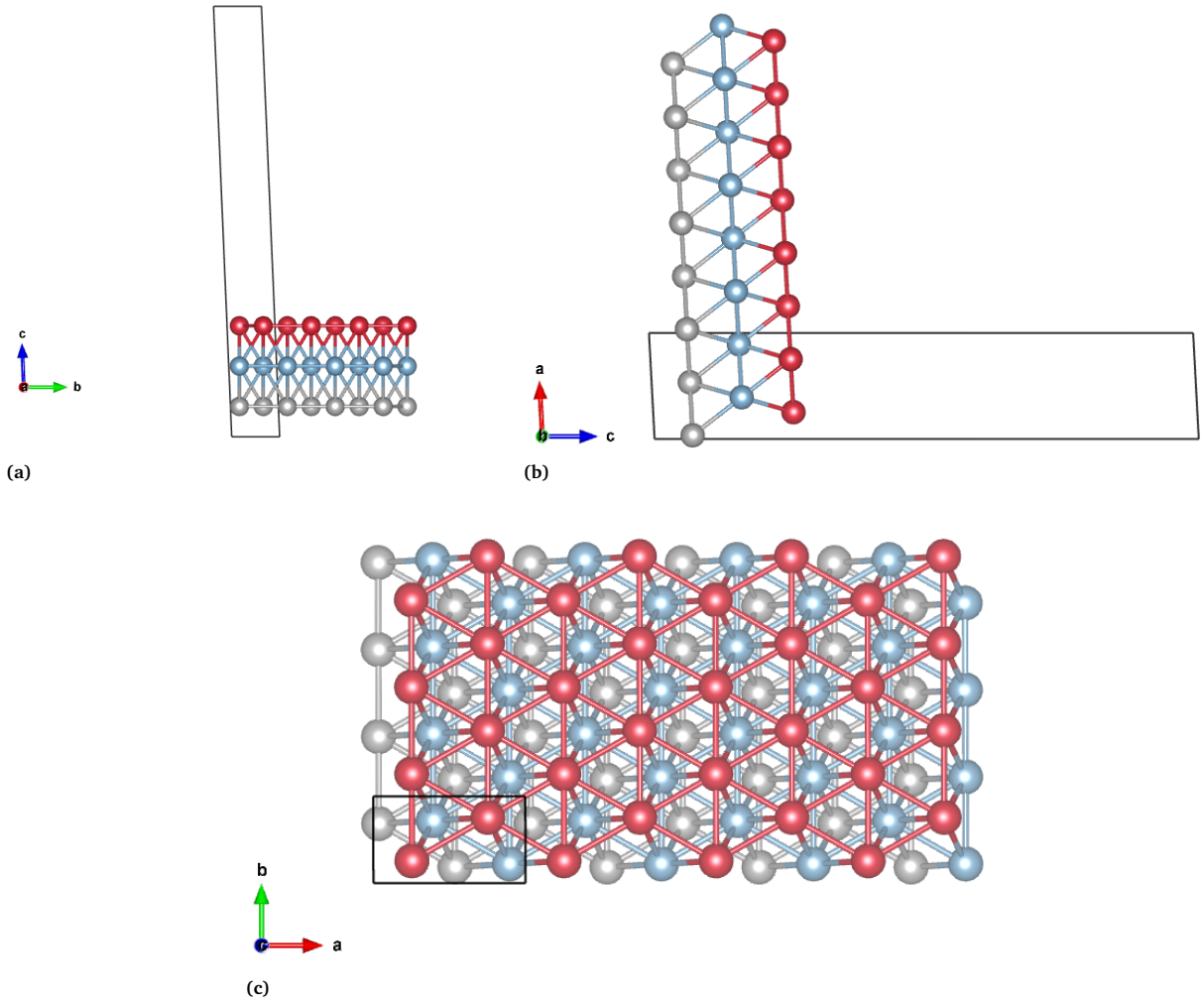


Fig. S2 Visualisations of the optimised trilayer structure aligned to unit cell axis. The bottom layer is shown in grey, the middle layer in blue, and the top layer in red.

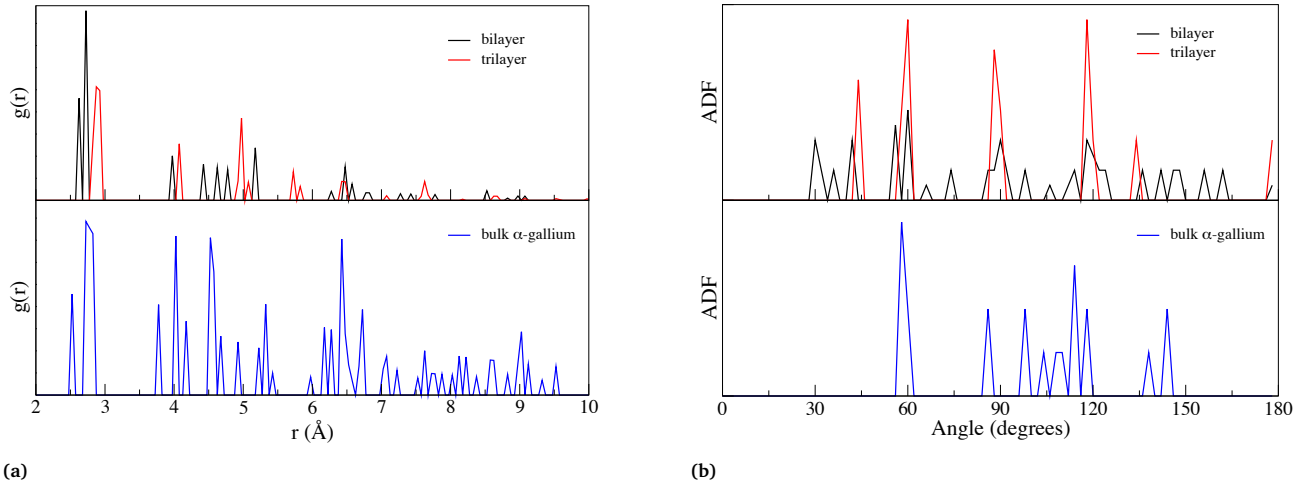


Fig. S3 (a) Radial distribution function, g(r), and (b) angular distribution function (ADF) for the lowest energy, optimised bilayer and trilayer structures. For the ADF, we include only the angles between atoms within the first coordination shell. For both the radial distribution function and ADF, we include an analysis of bulk α-gallium optimised with the same computational settings (PBEsol, PAW) for comparison.

3 Band structure and phonon dispersion

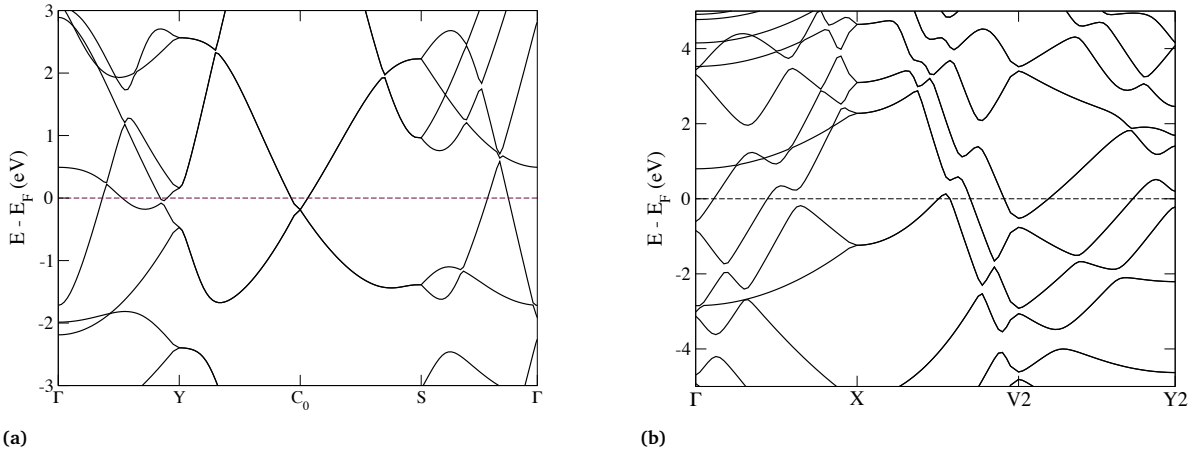


Fig. S4 Band structures for the lowest energy, optimised structures of (a) bilayer and (b) trilayer gallenene.

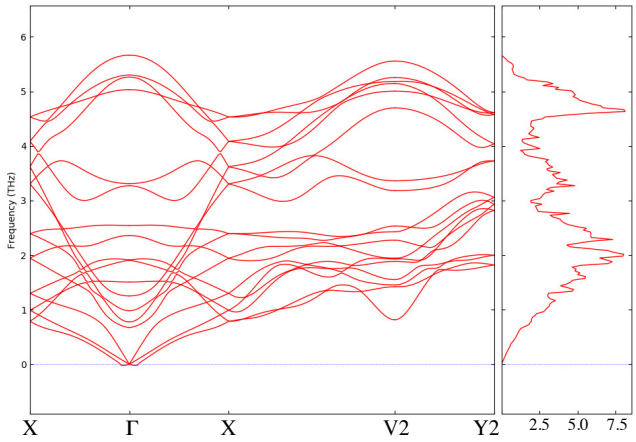


Fig. S5 The phonon dispersion for the lowest energy, optimised trilayer gallenene structures, calculated using a $4 \times 7 \times 1$ unit cell (168 gallium atoms), illustrating that the optimised trilayer is dynamically stable. The calculation yielded very small imaginary frequencies near the gamma point; however, the eigenvectors for these frequencies show that they are pure translations (all atoms move the same amount, in the same direction) and we have further verified that they arise solely due to numerical noise. As can be seen by the figure, the magnitudes of the frequencies are also negligible. Note: for presentation purposes, we have included the path from X-Γ (left), which is not included in the band structure of Fig. S4b.