

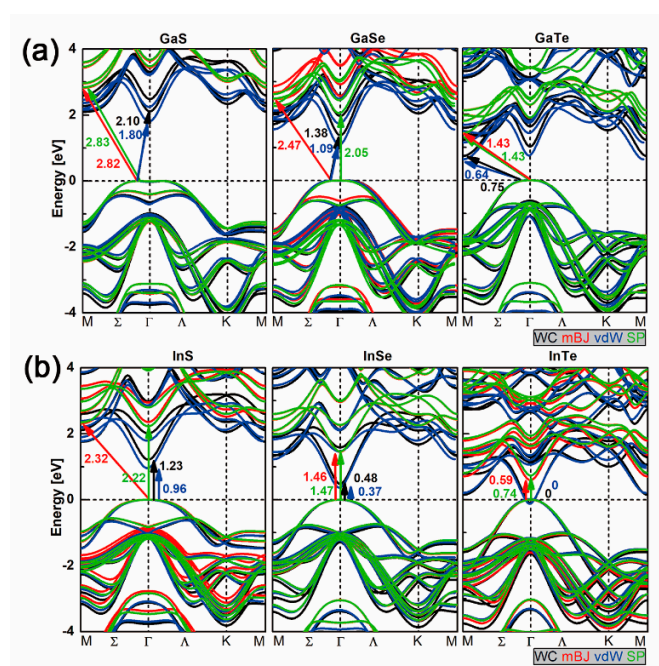
# Supplement Materials: Investigation on structure-property relationships of 2D Ga/In chalcogenides

Pingping Jiang <sup>1</sup>, Pascal Boulet <sup>1,\*</sup> and Marie-Christine Record <sup>2</sup>

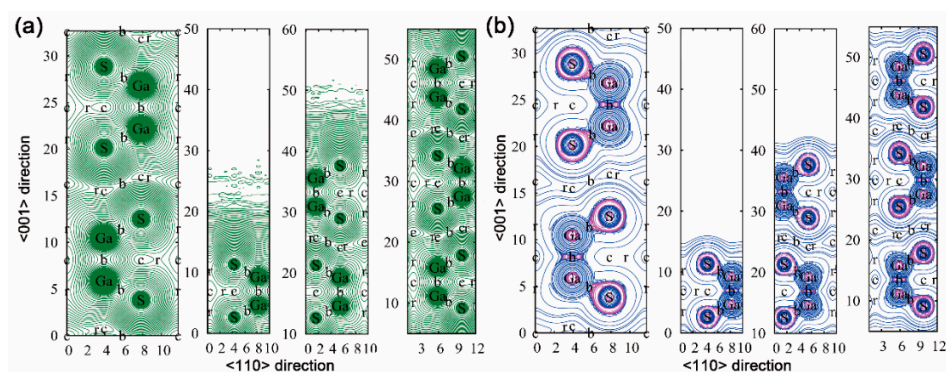
<sup>1</sup> Aix-Marseille Univ., CNRS, MADIREL, 13013 Marseille, France; pingping.jiang@etu.univ-amu.fr

<sup>2</sup> Aix-Marseille Univ., Université de Toulon, CNRS, IM2NP, 13013 Marseille, France; m-c.record@univ-amu.fr

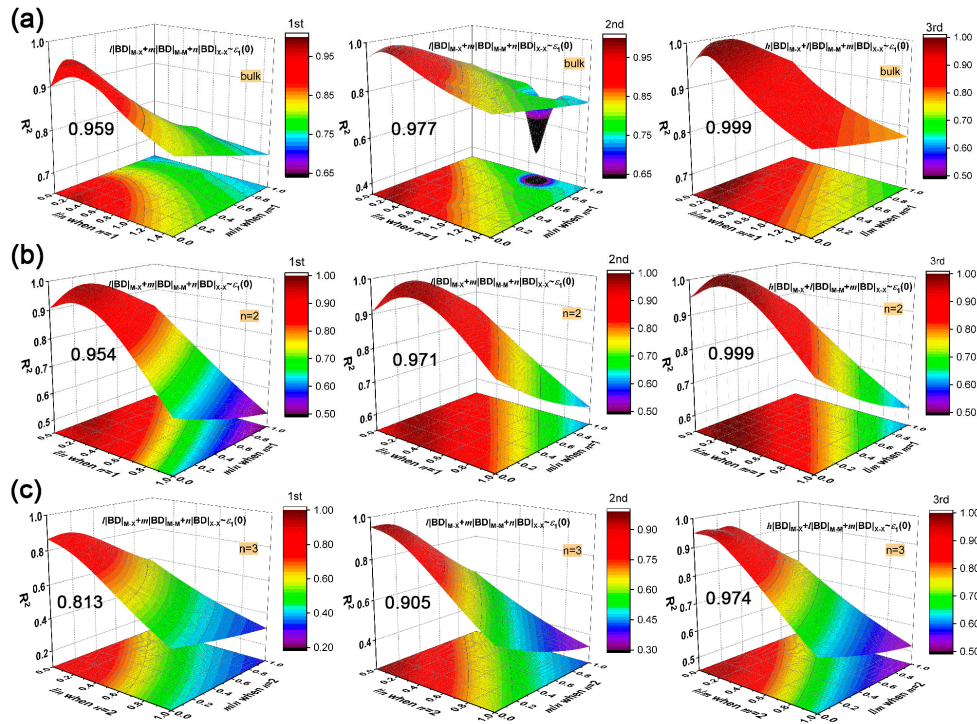
\* Correspondence: pascal.boulet@univ-amu.fr; Tel: +33-413-55-18-10



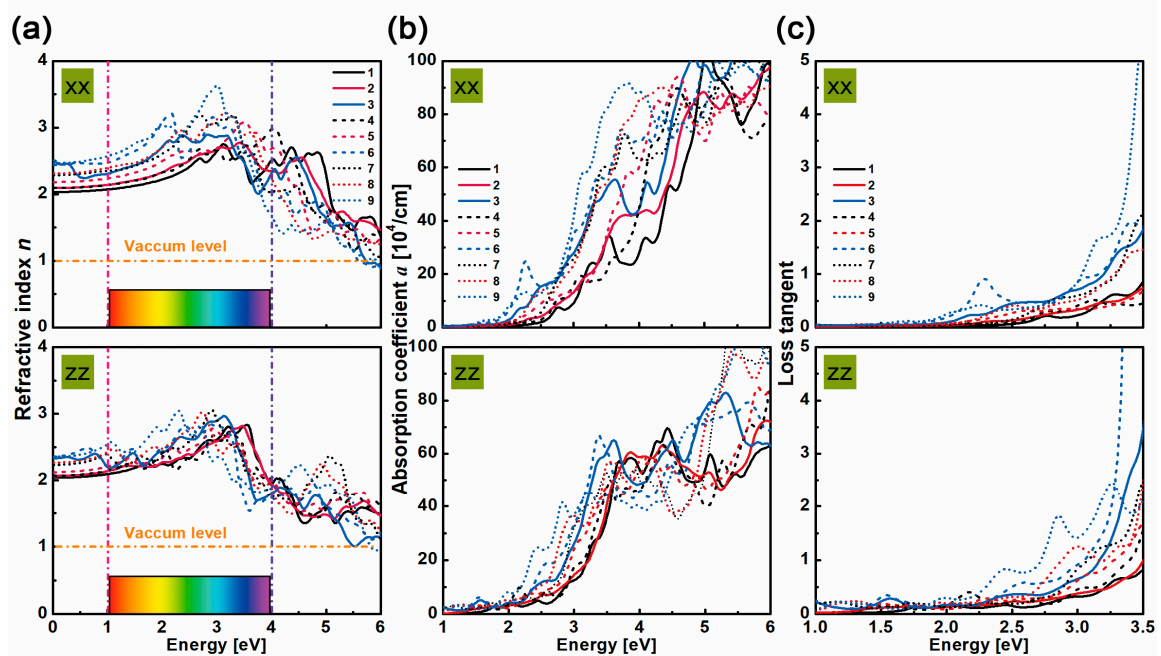
**Figure S1.** Band structures of bulk GaX (a) and InX (b) (X = S, Se, Te) calculated using WC-GGA with (green curves) and without polarization (black curves), mBJ (red curves) and optB88-vdW (blue curves) functionals.



**Figure S2.** Electron density (a) and Laplacian (b) distributions of bulk, mono-, bi, and trilayered GaS in (-110) plane. Labels "b", "r" and "c" represent bond, ring and cage critical points at the zero-flux surface respectively.



**Figure S3.** Polynomial fitting  $h|BD|_{M-X} + l|BD|_{M-M} + m|BD|_{X-X}$  vs.  $\epsilon_1(0)$  as equation order goes from the first to second and to third: Coefficient of determination  $R^2$  of bulk (a), bilayer (b), trilayer (c) by adjusting  $h/m$  and  $l/m$  ratios and their respective maximum  $R^2$ .



**Figure S4.** In-plane (xx) and out-of-plane (zz) refractive indexes (a), absorption coefficients (b) and loss tangents (c) of  $N^1-9$  GaX/InX.



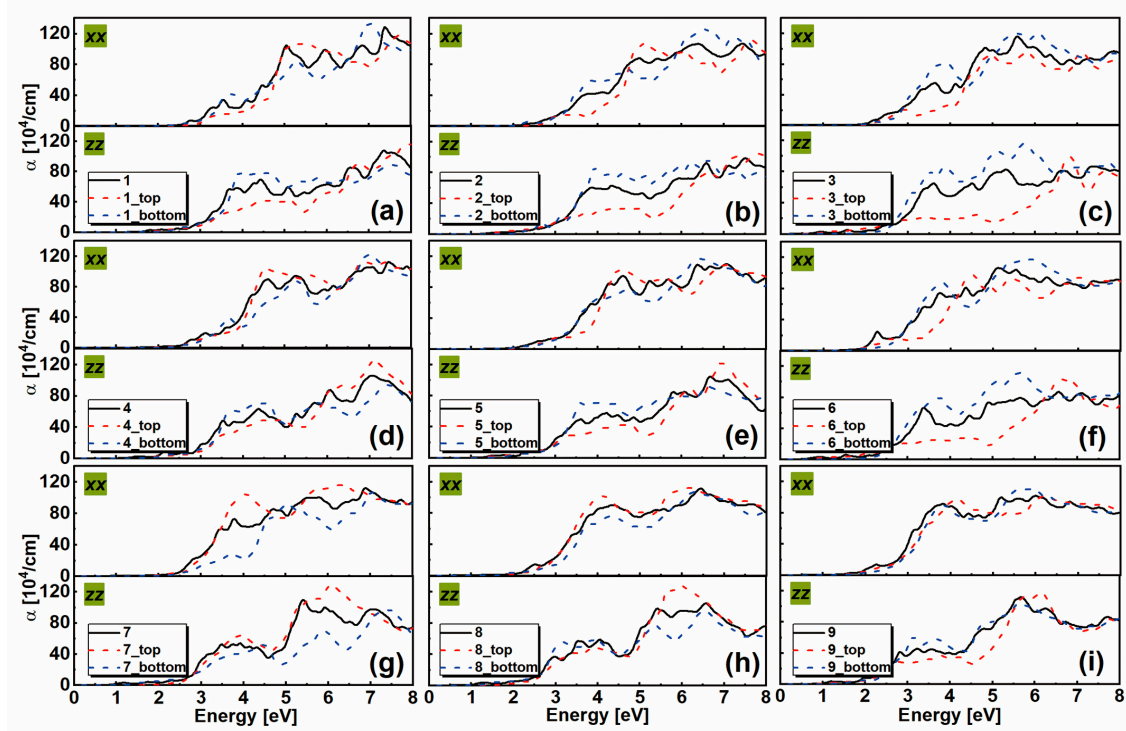


Figure S5. In-plane ( $xx$ ) and out-of-plane ( $zz$ ) absorption coefficients of  $N^o1-9$  GaX/InX compared with those of their constitutive top and bottom bilayers (a)–(i).

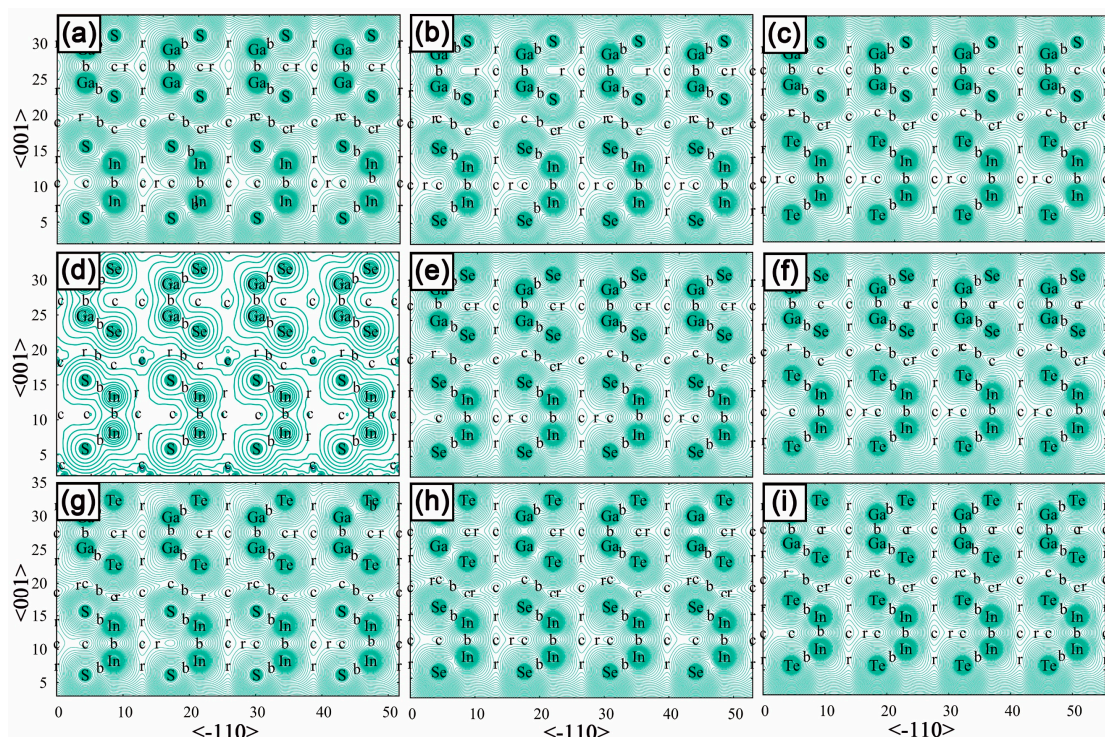


Figure S6. Electron density distributions of  $N^o1-9$  GaX/InX along the  $\langle 110 \rangle$  plane (a)–(i). Labels “b”, “r” and “c” represent bond, ring and cage critical points at zero-flux surface respectively.



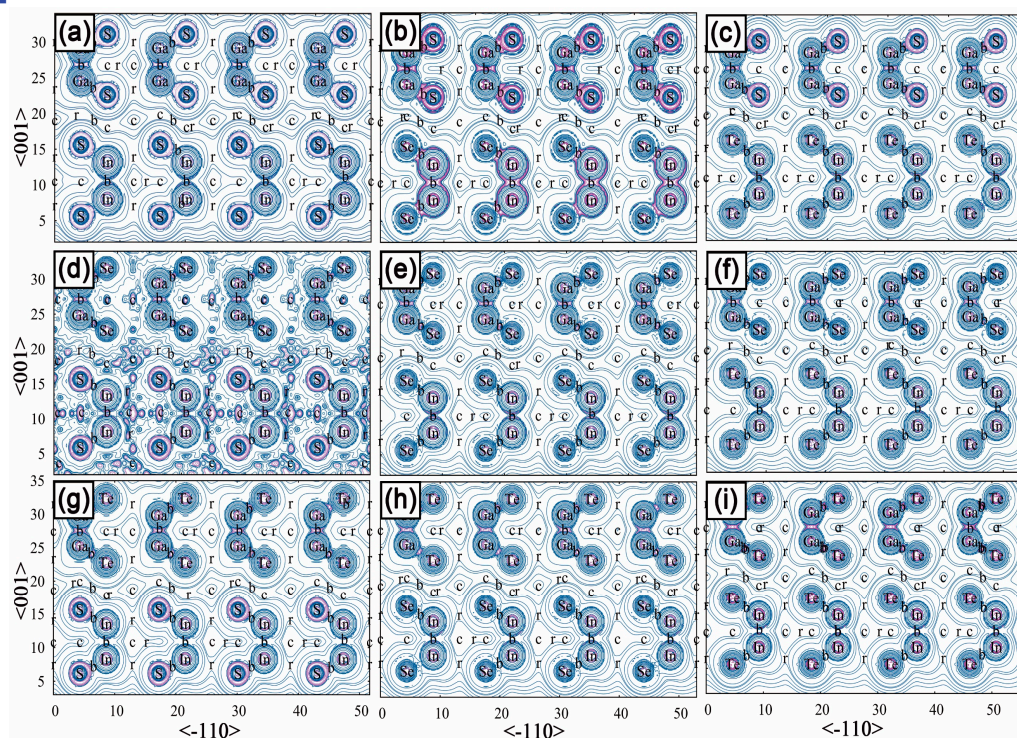


Figure S7. Laplacian of electron density distributions of N°1–9 GaX/InX along the  $\langle 110 \rangle$  plane (a–i). Labels “b”, “r” and “c” represent bond, ring and cage critical points at zero-flux surface respectively.

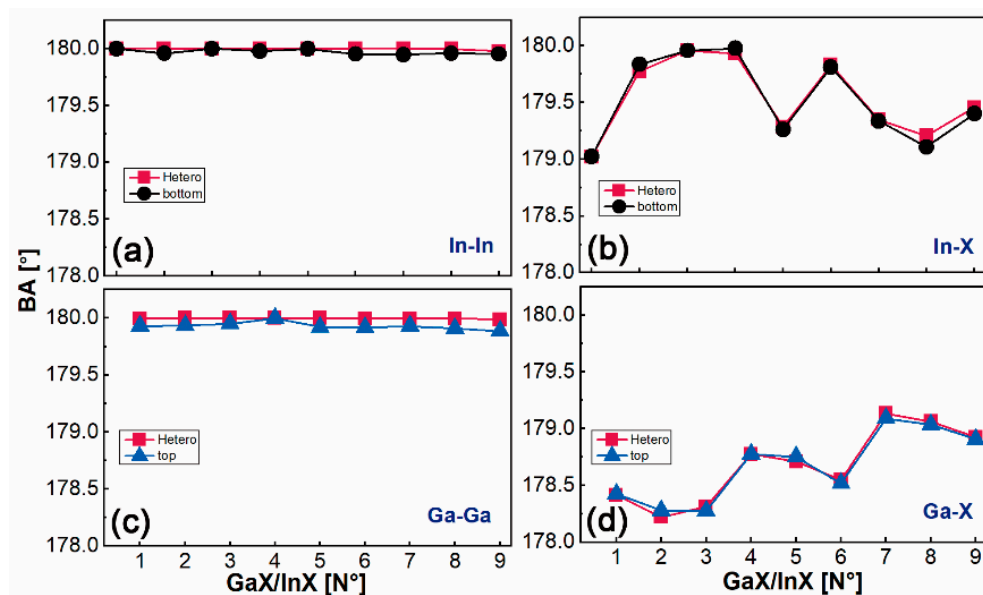


Figure S8. Bond angle (BAs) of In–In (a), In–X (b), Ga–Ga (c) and Ga–X (d) bonds of N°1–9 GaX/InX (red color), and the constitutive top  $[\text{GaX}]_2$  (blue color) and bottom  $[\text{InX}]_2$  (black color).