Supporting Information for

GeC/GaN vdW Heterojunctions: a Promising Photocatalyst for Overall Water Splitting and Solar Energy Conversion

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Figure S1: (Color online) The total energy per unit cell as a function of lattice constant for (a) GeC monolayer, (b) GaN monolayer, and six stacked GaN/GeC vdW heterostructures with (c) AB1 stacking, (d) AB2 stacking, (e) AB3 stacking, (f) AB4 stacking, (g) AB5 stacking, and (h) AB6 stacking, where $a_0$ is the equilibrium lattice constant.

Figure S2: (Color online) Layer-resolved atom-projected band structures calculated by PBE for (a) GeC monolayer, (b) GaN monolayer, and six stacked GaN/GeC vdW heterostructures with (c) AB1 stacking, (d) AB2 stacking, (e) AB3 stacking, (f) AB4 stacking, (g) AB5 stacking, and (h) AB6 stacking. In (c)-(h), blue marks are contributed by GaN layer of GeC/GaN vdW heterostructures, and red marks are contributed by GeC layer of GeC/GaN vdW heterostructures.
Empirical Band Alignment

Figure S3: (Color online) Band Alignment calculated by an empirical formula ($E_{\text{CBM/VBM}} = -X \pm 0.5E_g^{GW}$), where the vacuum level is set to zero.

Figure S4: (Color online) (a) Band Alignment calculated by an empirical formula (reds and blues respectively displaying the results of $GW$ and HSE06), where the vacuum level is set to zero. (b) Band gaps for PBE (blacks), HSE06 (blues), and $GW$ (reds).
Figure S5: (Color online) Band Alignment calculated by a first principles DFT scheme $(E_{\text{CBM/VBM}} = E_{\text{BGC}} \pm 0.5E_{\text{GW}}^0)$ for (a) GeC monolayer, (b) GaN monolayer, and six stacked GaN/GeC vdW heterostructures with (c) AB1 stacking, (d) AB2 stacking, (e) AB3 stacking, (f) AB4 stacking, (g) AB5 stacking, and (h) AB6 stacking. Horizontal red and blue dashed lines represent the vacuum levels for GeC layer and GaN layer, respectively. $\Delta V$ is the potential difference between the vacuum levels of GeC layer and GaN layer. $E_{\text{int}}$ represents the intrinsic electric field of the GeC/GaN vdW heterostructures materials.

Figure S6: (Color online) Band Alignment calculated by a first principles DFT scheme (HSE06) for (a) GeC monolayer, (b) GaN monolayer, and six stacked GaN/GeC vdW heterostructures with (c) AB1 stacking, (d) AB2 stacking, (e) AB3 stacking, (f) AB4 stacking, (g) AB5 stacking, and (h) AB6 stacking. Horizontal red and blue dashed lines represent the vacuum levels for GeC layer and GaN layer, respectively. $\Delta V$ is the potential difference between the vacuum levels of GeC layer and GaN layer. $E_{\text{int}}$ represents the intrinsic electric field of the GeC/GaN vdW heterostructures materials.
Figure S7: (Color online) (a) Optical absorption coefficient $\alpha(\omega)$ for GaN monolayer, GeC monolayer, and GeC/GaN vdW heterostructures with electron-hole interaction (red lines) and without electron-hole interaction (blue lines), where $\omega$ is photon energy. (b) Exciton binding energies for GaN monolayer, GeC monolayer, and GeC/GaN vdW heterostructures.

Figure S8: (Color online) Side and top views of electron probability distributions of the lowest lying optically bright excitons for (a) GeC monolayer, (b) GaN monolayer, and six stacked GaN/GeC vdW heterostructures with (c) AB1 stacking, (d) AB2 stacking, (e) AB3 stacking, (f) AB4 stacking, (g) AB5 stacking, and (h) AB6 stacking. Blue points make the position of the hole.
Figure S9: (Color online) GeC/GaN vdW heterostructure with AB1 stacking of top views of geometric structures at $t = 0$ and 3000 fs. Energy (blue) and temperature (red) as a function of quantum molecular dynamics (QMD) simulations time ($t$) at 300 K in a $4 \times 4 \times 1$ supercell.
Figure S10: (Color online) GeC/GaN vdW heterostructure with AB5 stacking of top views of geometric structures at \( t = 0 \) and 3000 fs. Energy (blue) and temperature (red) as a function of quantum molecular dynamics (QMD) simulations time \( (t) \) at 300 K in a \( 4 \times 4 \times 1 \) supercell.