

Enhancing AsH₃ gas sensing properties via transition metal doping in divacancy graphene: a first-principles investigation

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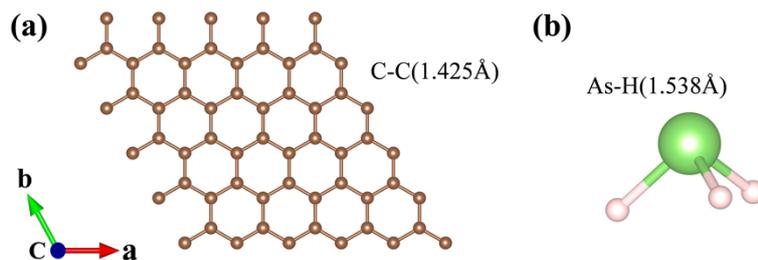


Fig. S1 (a) Ball-and-stick model of the optimized graphene structure; (b) ball-and-stick model of the optimized AsH₃ molecule. Brown, green, and light pink spheres represent C, As, and H atoms, respectively.

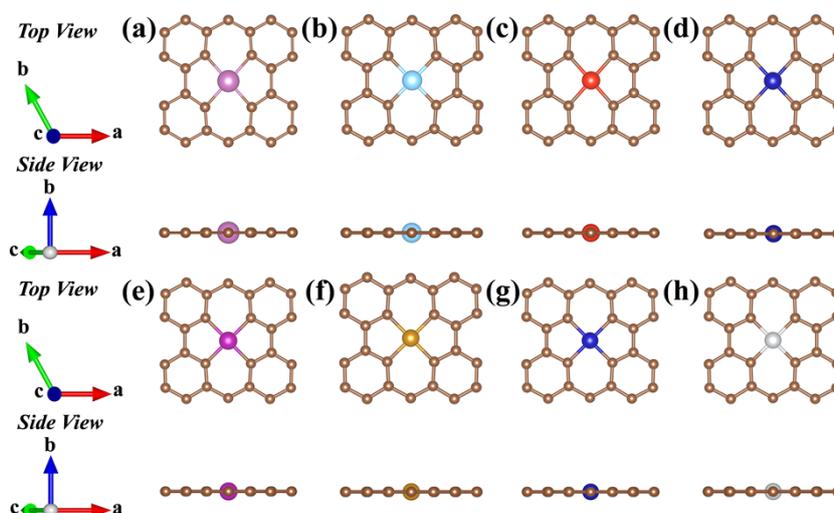


Fig. S2 Ball-and-stick models of optimized TM-DVG. (a-h) correspond to Sc-DVG, Ti-DVG, V-DVG, Cr-DVG, Mn-DVG, Fe-DVG, Co-DVG, and Ni-DVG, respectively. Brown, light purple, light blue, red, blue, purple, gold, orange, and silver spheres represent C, Sc, Ti, V, Cr, Mn, Fe, Co, and Ni atoms, respectively.

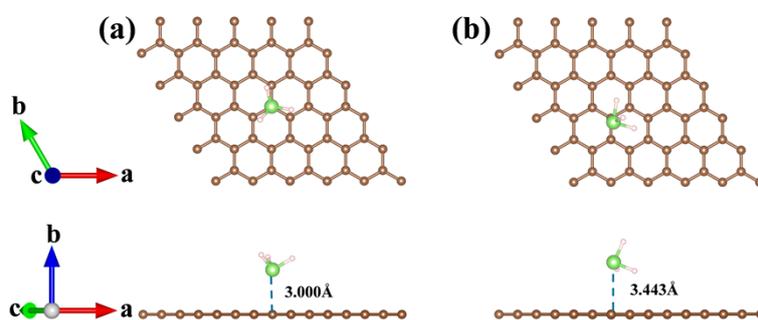


Fig. S3 (a) Initial configuration of AsH₃ adsorbed on pristine graphene. (b) Optimized configuration of AsH₃ adsorbed on pristine graphene.

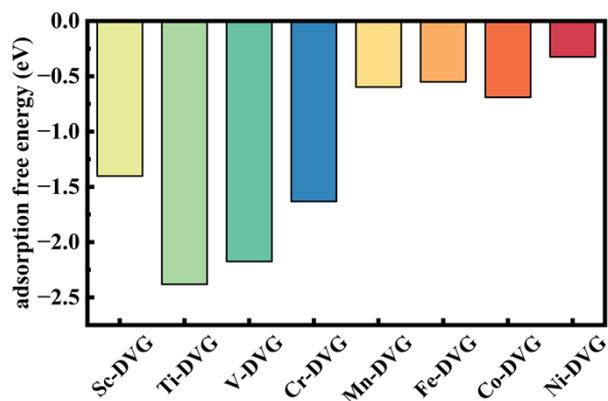


Fig. S4 The ΔG_{ads} of AsH_3 molecules on TM-DVG (TM = Sc, Ti, V, Cr, Mn, Fe, Co, Ni) at 300 K.

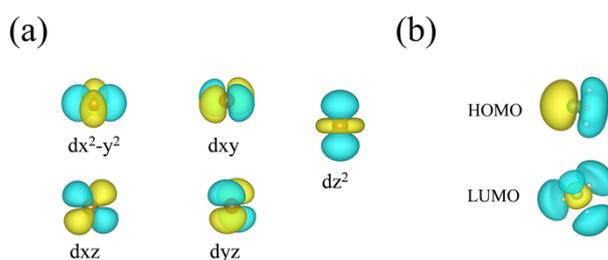


Fig. S5 (a) Five d-orbital configurations of the TM atom. (b) HOMO/LUMO configurations of the isolated AsH_3 molecule.

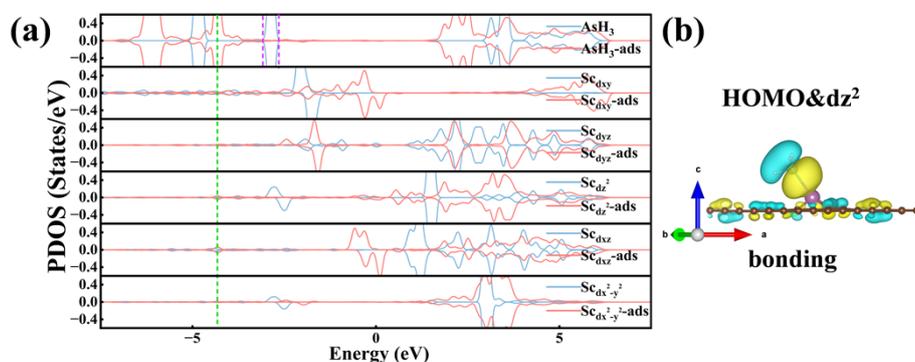


Fig. S6 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Sc and AsH_3 before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH_3 is highlighted within the purple box. (b) Partial charge density (PCD) with an isosurface value of $1 \times 10^{-8} \text{ e} \cdot \text{\AA}^{-3}$.

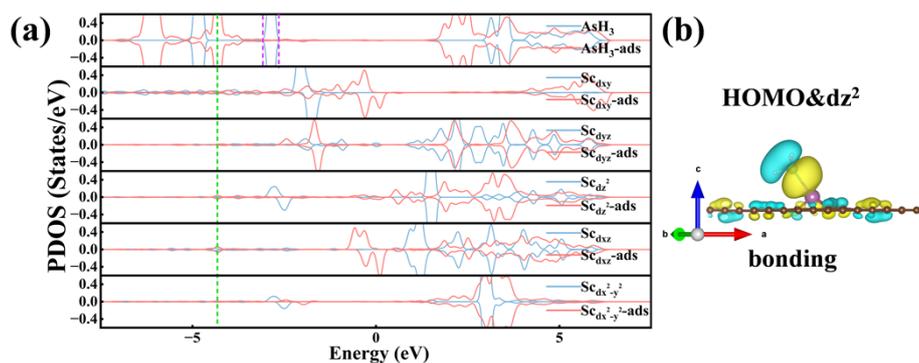


Fig. S7 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Ti and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with an isosurface value of $1 \times 10^{-8} e \cdot \text{\AA}^{-3}$.

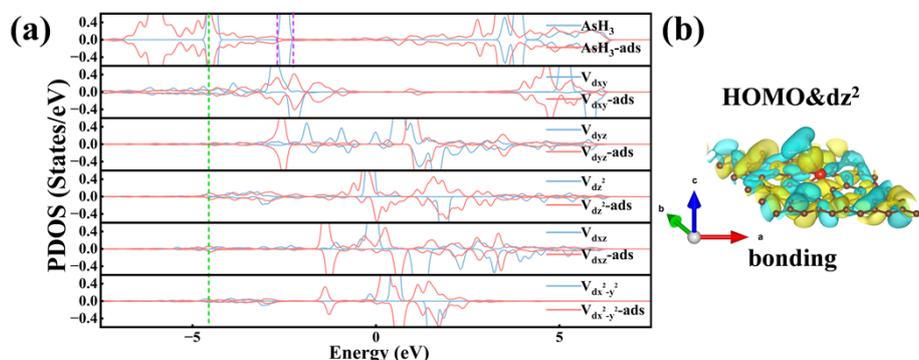


Fig. S8 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal V and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with an isosurface value of $1 \times 10^{-8} e \cdot \text{\AA}^{-3}$.

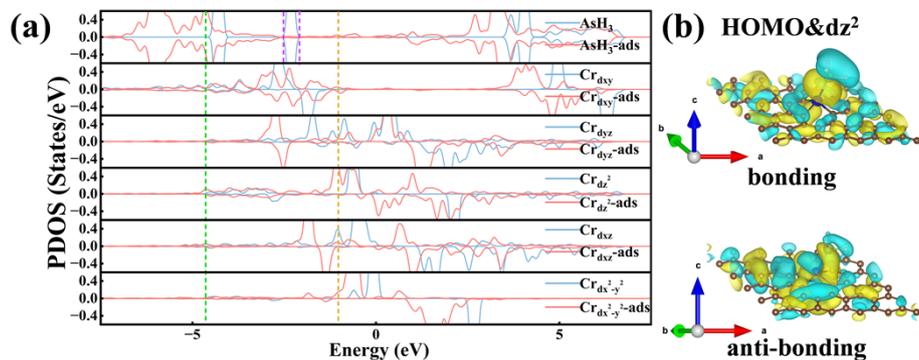


Fig. S9 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Cr and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with an isosurface value of $1 \times 10^{-8} e \cdot \text{\AA}^{-3}$ and $9.5 \times 10^{-9} e \cdot \text{\AA}^{-3}$.

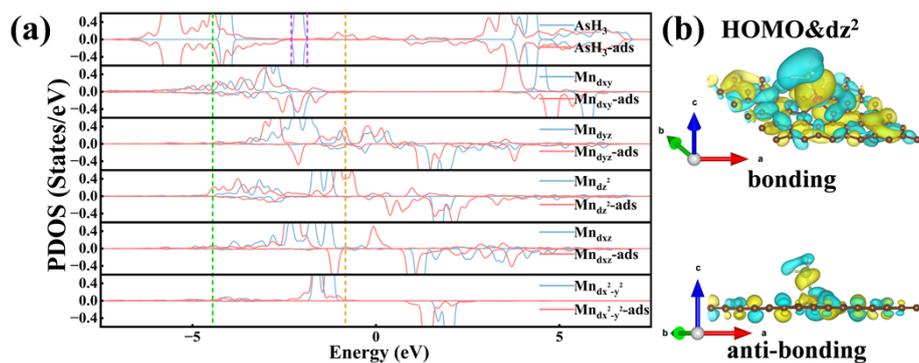


Fig. S10 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Mn and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with isosurface values of $1 \times 10^{-8} \text{ e} \cdot \text{\AA}^{-3}$ and $2.8 \times 10^{-8} \text{ e} \cdot \text{\AA}^{-3}$.

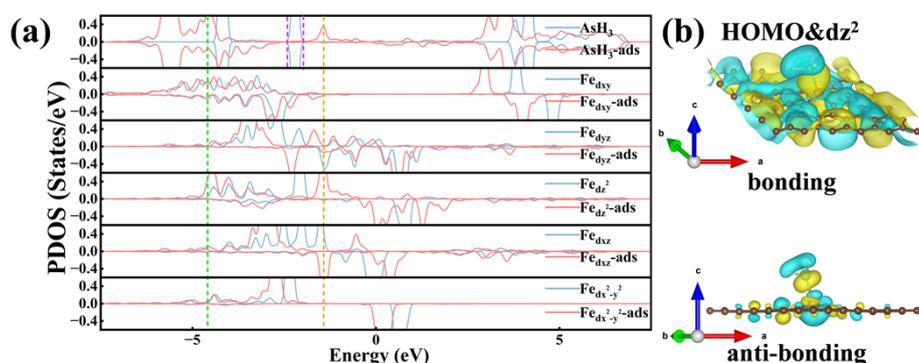


Fig. S11 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Fe and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with isosurface values of $7 \times 10^{-9} \text{ e} \cdot \text{\AA}^{-3}$ and $2.8 \times 10^{-8} \text{ e} \cdot \text{\AA}^{-3}$.

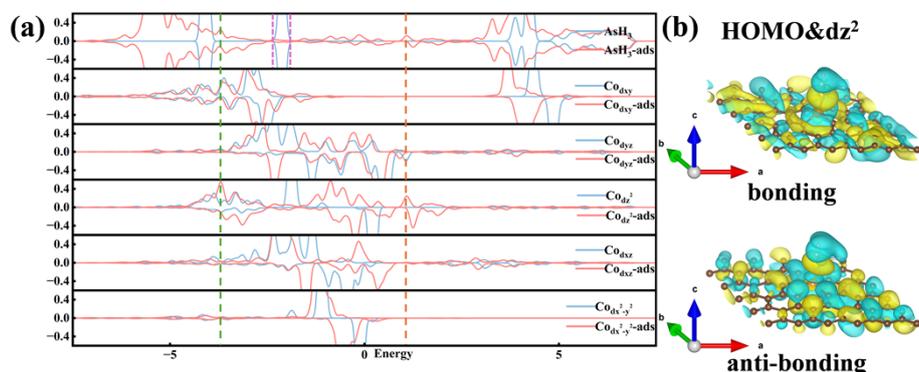


Fig. S12 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Co and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density

(PCD) with isosurface values of $1 \times 10^{-8} e \cdot \text{\AA}^{-3}$ and $1.5 \times 10^{-8} e \cdot \text{\AA}^{-3}$.

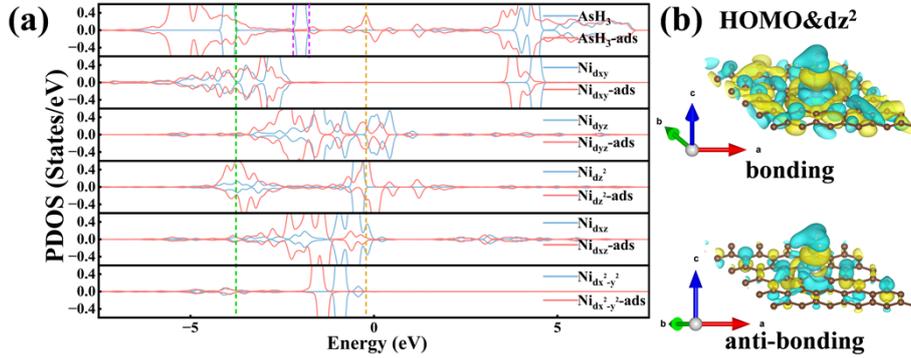


Fig. S13 (a) Projected density of states (PDOS) of the five d-orbitals of the transition metal Ni and AsH₃ before (blue) and after (red) adsorption. Positive PDOS values correspond to spin-up states, while negative values represent spin-down states. The HOMO of isolated AsH₃ is highlighted within the purple box. (b) Partial charge density (PCD) with isosurface values of $9 \times 10^{-9} e \cdot \text{\AA}^{-3}$ and $1.5 \times 10^{-8} e \cdot \text{\AA}^{-3}$.

Table S1 Formation energy (E_f) and charge transfer amount (Δq) of the optimized TM-DVG structures, and the distance between the TM atoms and adjacent C atoms ($d_{\text{TM-C}}$). Negative values of Δq indicate charge loss from the TM.

	E_f (eV)	Δq (e)	$d_{\text{TM-C}}$ (Å)
Sc-DVG	-0.662	-1.582	1.947
Ti-DVG	-0.570	-1.787	2.013
V-DVG	-0.082	-1.305	1.987
Cr-DVG	-0.498	-1.126	1.976
Mn-DVG	-1.856	-1.223	1.954
Fe-DVG	-1.473	-1.094	1.926
Co-DVG	-1.347	-0.854	1.900
Ni-DVG	-2.075	-0.634	1.880

Table S2 Theoretical Desorption Time (unit: s) of AsH₃ on TM-DVG Surface at Various Temperatures.

AsH ₃	τ (300K)	τ (400K)	τ (500K)
Sc-DVG	4.75E+15	3.22E+08	1.61E+04
Ti-DVG	1.64E+33	4.58E+21	5.36E+14
V-DVG	5.90E+29	1.20E+19	4.60E+12
Cr-DVG	1.15E+20	6.25E+11	6.86E+06
Mn-DVG	1.97E+03	1.67E-01	5.99E-04
Fe-DVG	4.20E+02	5.22E-02	2.37E-04
Co-DVG	1.62E+05	4.55E+00	8.44E-03
Ni-DVG	8.14E-02	8.57E-05	1.40E-06