SUPPLEMENTARY INFORMATION

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Electronic Properties of Nanodiamond Decorated Graphene

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Figures



Figure S1. Additional electron microscopy observation of ND/G hybridized structure. a, An optical microscopy image of homogeneously dispersed F-NDs in PMMA solution. **b**, SEM image of ND/G film on copper film after the CVD growth. Scale bar=1µm. **c**, HRTEM image of graphene film decorated by ND in low magnification. Scale bar=500nm. **d**, HRTEM image of ND on graphene in high magnification. Scale bar=100nm.



Figure S2. Controlling density of NDs on graphene. SEM images of ND dispersed on graphene film at different density by different speed of spin coating on copper foil. a, 500 rpm. b, 1000 rpm. c, 1500 rpm. d, 2000 rpm. e, 2500 rpm. f, 3000 rpm. Scale bar=1μm.



Figure S3. Analysis of the interface of ND/G film by HRTEM characterizations. a, HRTEM images of the hybrid nanostructure at interface. Scale bar is 2nm. The inset is SAED pattern in the graphene region without ND, showing high quality of graphene film. Scale bar is $5nm^{-1}$. **b**, A

statistical distribution of the ND/G cluster sizes. The majority of clusters size range from 40 to 60 nm. **c**, A comparison of EELS data of graphene film without and with ND, respectively. **d**, The graphene film with ND particles was mapped by EELS (right). Scale bar is 10nm.



Figure S4. Raman spectra of ND/G with different ND density. **a**, Raman spectra of ND/G with different ND density. (a) Raman comparison of the ND/G film with different ND density showing an increase in the intensity of both D (~1346 cm⁻¹) and D' (~1620 cm⁻¹) peaks with increasing ND density. **b**, A plot of the ratio of the intensity of G to D peak as function of speed of spin coating on copper foil. This indicates that the ratio of sp^2 to sp^3 in ND/G film decreases with increasing ND density.



Figure S5. XPS comparison of F-ND and ND/G. It has been found that C–F bond has been broken after the CVD growth of ND/G film in 1000 °C for 20 min. From the XPS measurement, we may safely exclude the effect of fluorine atom in the interface of ND/G hybridized molecular structure.



Figure S6. Proton-Induced X-ray Emission (PIXE) scan for two samples with 2 MeV proton beam line. Carbon, the dominant surface material, is outside the detection range of PIXE.



Figure S7. Low field MR vs. (parallel) magnetic field for graphene/ND sample in twoterminal geometry. [Inset: R vs. V_{BG} at T= 10K, B = 0 T]



Figure S8. a, Conductance vs. V_{SD} at T = 2.5 K, V_{BG} = 9 V, for graphene nanoconstriction GNC-1 discussed in main text. [Inset: Same plot from a separate sweep in the vicinity of low bias. **b**, Conductance vs. V_{BG} for GNC-1 at T = 2.5 K and a low-bias of V_{SD} = 1.43 mV. $\Delta VBG \sim$ 9 V is the width of conductance suppression along the gate axis.



Figure S9. a, Resistance vs. V_{BG} - V_{NP} at different temperatures for graphene nano-constriction GNC-2. **b**, Resistance vs. V_{BG} - V_{NP} at different temperatures for graphene nano-constriction GNC-3. **c**, Resistance vs. V_{BG} - V_{NP} at T = 2.5 K and B = 0 T (red), B = 8 T (black) for graphene nano-constriction GNC-2. **d**, Resistance vs. V_{BG} - V_{NP} at T = 2.5 K and B = 0 T (red), B = 8 T (black) for graphene nano-constriction GNC-3. **d**, Resistance vs. V_{BG} - V_{NP} at T = 2.5 K and B = 0 T (red), B = 8 T (black) for graphene nano-constriction GNC-3. Note the MR in (c), (d) is measured with a high-bias current (I = 10 nA).



Figure S10. MR vs. B at T = 10 K for pristine CVD graphene (red) and ND/graphene (blue), both gated at charge neutrality point.



Figure S11. MR vs. B for pristine CVD graphene on oxygen terminated single-crystal diamond substrate (blue) at T = 25 K, $n \sim 2 \times 10^{12}/cm^2$ and ND/graphene on SiOx substrate (green) at T = 20 K, $n \sim 1.8 \times 10^{12}/cm^2$.

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