Supporting Information:

Screening of Hydrogen Bonding Interactions by a Single Layer Graphene

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Supporting Figures



Figure S1: Representative AFM topographic images of the bare (a, c) and graphene-coated (b, d) sapphire substrates using a 5 μ m x 5 μ m (a, b) and 10 μ m x 10 μ m (c, d) scan sizes. Scale bars indicated on the images. Similar RMS roughness for bare and graphene-coated sapphire substrates confirms conformal coating of graphene on the underlying sapphire substrate.



Figure S2: (a) Plots of strain energy release rate $(G, J/m^2, \text{ calculated by solving Equation 1 for each data point during loading and unloading) with respect to contact area <math>(m^2)$ for adhesion of PDMS lens to graphene-coated SiO₂/Si and bare SiO₂/Si. (b) Plots of G (J/m^2) with respect to contact area (m^2) for adhesion of PDMS to graphene-coated sapphire and bare-sapphire. Loading, open makers; unloading, filled markers.



Figure S3: (a)Adhesion measurements during approach (open markers) and retraction (solid markers), where a^3 vs. load (F) data is plotted for 0.7 MPa PDMS lenses on Gr/SiO₂/Si (pink circles) and SiO₂/Si (black triangles) substrates. (b) Comparison of mean W_a values calculated from loading (by fitting the loading data using Equation 1) and pull-off force (calculated using Equation 3) for uncoated and graphene-coated SiO₂/Si substrates. Error bars indicate ±1 standard deviation.



Figure S4: Adhesion measurements during approach (red open markers) and retraction (blue solid markers), where a^3 vs. load (F) data is plotted for PDMS on octadecyltrichlorosilane (OTS)-coated silicon wafer using 0.7 MPa (a) and 1.9 MPa (b) PDMS hemispherical lenses. The approach and retraction curves are fit with Equation 1 to obtain the work of adhesion (W_a) from loading and unloading (fit curves shown by solid lines).



Figure S5: SFG spectra collected using PPP polarization for air-Gr/sapphire under vacuum at room temperature (25°C, red open circles) and 150°C (blue open triangles). The solid lines represent fits obtained using the Lorentzian equation (Equation 4).



Figure S6: (a) Comparison of SFG spectra collected using PPP polarization for air-Gr/sapphire (red open circles) and PDMS-Gr/sapphire (blue open triangles) interfaces. (b) Comparison of PPP SFG spectra collected for air-sapphire (red open circles) and PDMS-sapphire (blue open triangles) interfaces. The solid lines in both (a) and (b) represent fits obtained using the Lorentzian equation (Equation 4).

Supporting Tables

Air-Sapphire	$\omega_q/\mathrm{cm}^{-1}$	3250	3592	3705			
	$\Gamma_q/{ m cm}^{-1}$	150	183	16			
	A_q	2	1945	175			
Air-Gr/Sapphire	$\omega_q/\mathrm{cm}^{-1}$	3301	3643	3702			
	$\Gamma_q/\mathrm{cm}^{-1}$	200	45	25			
	A_q	351	356	91			
PDMS-Sapphire	$\omega_q/\mathrm{cm}^{-1}$	2908	2968	3610	3689		
	$\Gamma_q/\mathrm{cm}^{-1}$	11	10	88	18		
	A_q	-50	153	3823	219		
PDMS-Gr/Sapphire	$\omega_q/\mathrm{cm}^{-1}$	2862	2909	2963	3000	3636	3708
	$\Gamma_q/\mathrm{cm}^{-1}$	10	14	14	34	50	23
	A_q	24	79	283	-319	1465	192

Table S1: SFG fitting parameters obtained by fitting the spectra shown in Figures 3 and S6.