Supporting Information

Evidence for Spin Glass Ordering Near the Weak to Strong Localization Transition in Hydrogenated Graphene

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Sample Raman spectra and estimate of defect free domain size L_a:

Raman spectroscopy was used to confirm the presence of single layer graphene by fitting the shape of the 2D peak (2679 cm^{-1}, second-order D mode), and to quantify the degree of hydrogenation using the I_D/I_G ratio, which is the relative intensity of the D mode (1345 cm^{-1}, appearing due to symmetry breaking at defect sites) with respect to the G mode (1588 cm^{-1}, E_{2g} phonon mode). All Raman spectra were taken under ambient conditions using a 514 nm laser excitation.

Figure S1 shows two sample Raman spectra of a graphene device before and after hydrogenation; this particular data set corresponds to the hydrogenated device with $I_D/I_G \sim 1.6$ as described in the main text. A rough estimate of the defect free domain size L_a before and after hydrogenation is found from $L_a^2(\text{nm}^2) = (1.8\times10^{-9})\lambda^4(I_G/I_D)$, where $\lambda = 514$ nm is the excitation wavelength.[SR1] For this particular data set shown in Figure S1 after hydrogenation and with I_D/I_G ~ 1.6 we find L_a ~ 8.9 nm, which is comparable to the value for the elastic mean free path L_e ~ 7.46 nm determined from the transport data.

Estimates of σ_S, n, and σ_D:

The short-range conductivity σ_S, the charge carrier density n at the Dirac point (DP), and the Drude conductivity σ_D are estimated from the measured transport of the graphene and hydrogenated graphene at zero magnetic field B. For example, Figure S2 shows the measured conductivity σ *versus* n at B = 0 T for one of our hydrogenated devices. Values for σ were obtained from $\sigma = L/RW$ where L and W are the sample length and width, respectively, and R is the sample resistance obtained by sourcing a current between electrodes 1 and 2, I_{12}, as labeled in Figure 1a of the main text and by measuring the resultant voltage drop along the current path using electrodes 3 and 4, V_{34}. Applying a back gate voltage V_g to the doped Si substrate allows us to vary n. We determine n from $n = 1/eR_H$ where e is the fundamental unit of electric charge and R_H is the Hall coefficient, defined as $R_H = R_{xy}/B$. R_{xy} was found from $R_{xy} = V_{78}/I_{12}$ (Figure 1a of the main text) with B = 2.6 T applied perpendicular to the plane of the graphene.

The data in Figure S2 for the hydrogenated graphene is well fitted by a diffusive transport model given by

$$\sigma(n) = \left(\frac{1}{ne\mu_L} + \rho_s + \rho_{ph}\right)^{-1} \qquad \text{(S1)}$$

where μ_L is the n-independent mobility due to long-range, uncorrelated charged impurity scattering, and ρ_s and ρ_{ph} are the contributions to the resistivity due to short-range scattering and acoustic phonon scattering, respectively. Here, μ_L and ρ_s are the free fitting parameters and ρ_{ph} = (0.1 Ω/K)×T.[SR2] From the data fitting with Equation S1 we can determine σ_S by $\sigma_S = 1/\rho_S$.

We estimate n at the DP using the full width at half maximum ΔW_{DP} of the $\rho = 1/\sigma$ vs. n peak. This results in an upper bound for the carrier density fluctuations δn_{max} due to charge carrier puddle formation near the DP. For example, for the data shown in Figure S2 we find $\delta n_{max} \sim 6.15 \times 10^{15}$ m^{-2}, which is comparable to that observed for exfoliated graphene on SiO_2.[SR3 – SR5] This value obtained here for δn_{max} indicates a scattering contribution from charged impurities at low n. Values for δn_{max} provide an estimate for n at the DP where contributions from charged impurity scattering dominates the transport.[SR6]

Lastly, using Matthiessen's Rule we estimate σ_D from $\sigma^{-1} = \sigma_S^{-1} + \sigma_D^{-1}$ where σ is the overall conductivity measured through transport measurements.

Interpreting the various length scales:

As an example, for the hydrogenated device with $I_D/I_G \sim 1.6$:

The elastic mean free path $L_e = \sigma_D h/\left[2e^2(\pi n)^{1/2}\right]$ is found to be $L_e \sim 7.46$ nm, which is comparable to the defect free domain size $L_a \sim 8.9$ nm (see section above titled: **Sample Raman spectra and estimate of defect free domain size *L*$_a$**). The magnetic length L_B is determined by $L_B = \left(\hbar/_{e\,B}\right)^{1/2} = {}^{26\,nm}/_{\sqrt{B}}$ and ranges from 58.1nm (at 0.2 T) to 16.1 nm at (2.6 T). The curved arc trajectory length r_C is determined by $r_C = {}^{m^*\,\mu}/_{e\,B}$, where m^* is the effective mass of the charge carriers in graphene and μ the carrier mobility found from the transport, which ranges from 87 fm (at 0.2 T) to 7 fm at (2.6 T). The thermal length L_T is determined by $L_T = \left(\hbar D / k_B T \right)^{1/2}$, where D is the diffusion constant (see the main text) and k_B is Boltzmann's constant, and is $L_T \sim 116$ nm and is independent of B. The cyclotron radius L_C is determined by $L_C = \hbar k_F / e B = \hbar (\pi n)^{1/2} / e B$, where k_F is the Fermi wave vector, which ranges from 457 nm (at 0.2 T) to 35.2 nm at (2.6 T). Additionally, the Fermi wavelength is $\lambda_F = 2\pi / k_F = 2\pi / (n \pi)^{1/2} \sim 45\ nm$. That λ_F is larger than L_a (the characteristic size of the system) suggests that quantum effects are important. However, if the transition to a linear MR for $B \sim B_S$ as described in the main text was governed by any of the calculated length scales then one may expect a transition of such a length scale from larger than to smaller than L_a when B is $\sim B_S$, yet such a transition is not observed to occur. Thus, we conclude that the transition to a negative linear MR at $B \sim B_S$ is not governed by any of these length scales.

Resistivity ρ *versus* V_g for varying B:

Plotted in Figure S3 is a sample trace of the resistivity ρ *versus* back gate voltage V_g taken at $T = 4.2$ K for $B = 0$ T (for both graphene and hydrogenated graphene) and $B = 2.6$ T (for the hydrogenated graphene). Figure S3 clearly shows that the largest change in the magnetoresistance occurs at the DP where the spin glass properties have been observed. As V_g is shifted to values above or below the DP, corresponding to higher electron and hole concentrations, respectively, the change in the magnetoresistance quickly tends to zero.

References:

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SR6. Matis, B. R.; Houston, B. H.; Baldwin, J. W. Influence of Spatial Inhomogeneity on Electronic and Magnetotransport in Graphene. *Phys. Rev. B* **2015,** 91, 205406.

Supporting Information figures:

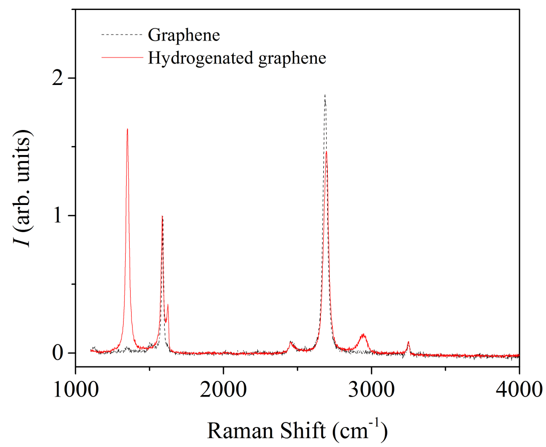

Figure S1. Sample Raman spectra of the initial graphene (dashed trace) and hydrogenated graphene (solid trace). The spectra have been normalized to the *G* mode intensity.

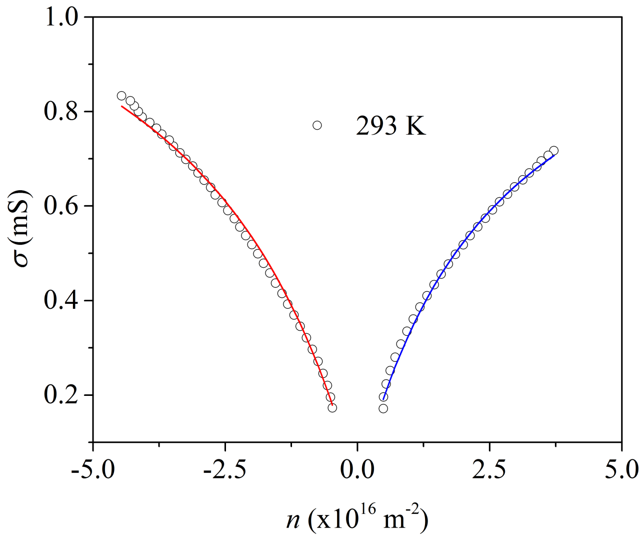

Figure S2. Sample trace of the conductivity σ *versus* charge carrier density n at magnetic field B = 0 T for hydrogenated graphene. The open circles are the experimental data and the solid lines are fits to Equation S1.

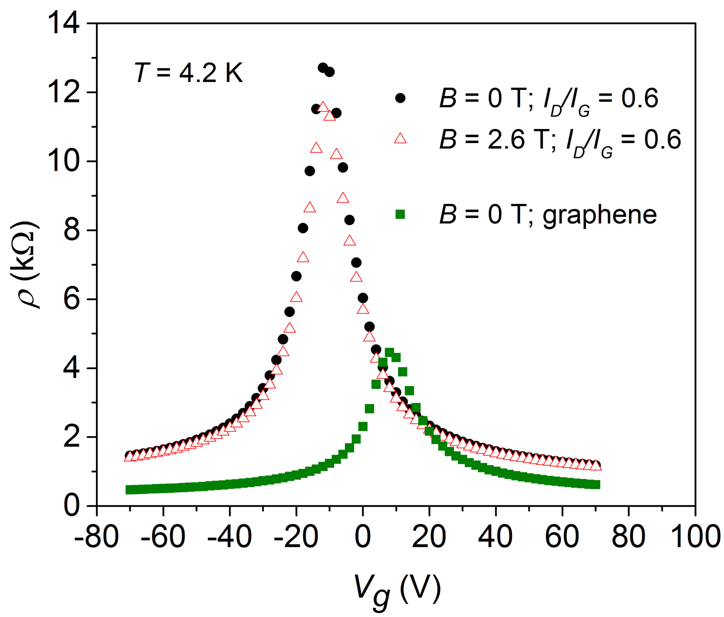

Figure S3. Sample traces of the resistivity ρ *versus* back gate voltage V_g for graphene (green trace) and hydrogenated graphene (black and red traces) at varying magnetic field B and for T = 4.2 K.

Table SI. Device characteristics for increasing I_D/I_G. The Ioffe-Regel parameter is given by $k\,L_e$.

I_D/I_G	ρ(B=0) (Ω)	ρ(B=0) ($h/2e^2$)	Measured spin glass	Localization regime	$k\,L_e$
0.6	11,470	0.89		WL	7.07
0.7	15,968	1.23	✓		5.08
0.8	17,072	1.32	✓		4.75
1.6	13,594	1.05	✓		5.97
2.0	71,597	5.55		SL	1.13