Annealing-induced magnetic moments detected by spin precession measurements in epitaxial graphene on SiC (Supplemental material)

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We report here on magnetotransport measurements of the reference sample before and after an annealing step and discuss the observability of a dip in the non-local in plane measurements.

The strong modification of the Hanle curves before and after annealing can be explained by either an enormous reduction of the diffusion constant after annealing or by the creation of localized moments, resulting in a strongly enhanced $g$-factor of the electrons. To check that annealing barely changes the transport properties, we characterized the reference sample with magnetotransport measurements before and after an annealing step at 150 °C for 15 minutes in vacuum. From these measurements we can extract the charge carrier density $n_s$ and the mobility $\mu$ in order to check the influence of annealing on these parameters. To this end we applied a perpendicular magnetic field and measured the Hall resistance $R_H$ at 1.7 K. We also determined the sheet resistance $R_s$ of graphene by applying the van der Pauw method. Finally, using the Drude formula, we calculated the carrier mobility $\mu = (R_s n_s e)^{-1}$. Both carrier density and mobility before and after annealing are given as insets in Fig. 1.

From this study we conclude that annealing can indeed change the charge carrier density and the sheet resistance slightly but the mobility is almost unaffected. That means that a change in the sheet resistance results mainly from a change in the charge carrier density. Taking the results from the reference sample we draw the following conclusion for the sample in which spin transport was studied. In this the sheet resistance increased slightly upon annealing which can be now ascribed to a minute reduction in the charge carrier density. Therefore the charge diffusion constant for our spin transport sample is only slightly decreased as $D_s \propto \sqrt{n_s}$ and can not explain the strong reduction of the spin diffusion constant extracted from the Hanle data when using the $g$-factor for a free electron.

We also performed Hall measurements for a number of temperatures after the annealing step, and determined the mobility $\mu$ at $T = 1.7 \ldots 40$ K. From those $T$-dependent measurements we conclude that the carrier density $n_s$ is barely $T$-dependent but the mobility increases as the temperature is lowered (Fig. 2). This information, together with the $T$-dependence of the sheet resistance of the spin transport sample allows us to model the $T$-dependence of the diffusion constant, which we need to determine the $T$-dependence of the $g$-factor.

Furthermore we discuss the dip feature around $B = 0$ in the non-local spin valve measurements which was experimentally observed in Ref. 1. Kawakami et al. observe a pronounced dip in the non-local resistance when they sweep the magnetic field in the direction along the ferromagnetic electrodes. This feature can be explained assuming the effective exchange-field model where the spin

FIG. 1. Magnetotransport measurement of the reference sample before and after an annealing step.

FIG. 2. Charge carrier density $n_s$ and mobility $\mu$ versus temperature $T$ of the reference sample.
The spin-lifetime \( \tau^{\text{so}} \) obtained from non-local Hanle measurements is quite long so an ensemble of fluctuating spins with \( \Delta B = 6.78 \) mT and \( \tau_c = 192 \) ps leads to a sizeable reduction of \( T_1^{\text{total}} \).

Since the spin-flip length is \( L_s = \sqrt{D_s T_1^{\text{total}}} \) and the non-local signal in the tunneling regime\(^{2,3} \) depends on \( L_s \) according to

\[
R_{nl} = \frac{D^2 R_s L_s}{2W} \exp(-L/L_s),
\]

a variation of \( T_1^{\text{total}} \) directly modifies the observed non-local signal. In our case, the spin lifetime before and after annealing is 80 ps and 22 ps, respectively. Annealing creates localized moments, reducing the \( B \)-independent spin lifetime (modeled by \( \tau^{\text{so}} \) in Eq. (1)) due to spin-flip scattering with the localized moments, while the additional, \( B \)-dependent reduction resulting from the effective exchange field model is barely visible. This is shown in Fig. 3 and Fig. 4.

In Fig. 3 we reproduce the expected \( T_1^{\text{total}} \)-time for the experiment of the Kawakami group, showing a pronounced dip in \( T_1^{\text{total}} \) around \( B = 0 \). If we replace \( \tau^{\text{so}} \) with the experimental value for our sample after annealing, we find that for the same \( \Delta B = 6.78 \) mT and \( \tau_c = 192 \) ps as in Kawakami’s experiment the dip feature is now almost invisible (Fig. 4).

This also holds if we increase \( \Delta B \) to 20 mT and \( \tau_c \) to 3 ns (Fig. 5 and Fig. 6). The corresponding tiny change in \( R_{nl} \) of at most a few mΩ would be unobservable, given the noise level of about 10 mΩ. Indeed, experimentally we did not observe such a feature.

![Fig. 3. \( T_1^{\text{total}} \) from Kawakami’s group.](image)

![Fig. 4. \( T_1^{\text{total}} \) with \( \tau^{\text{so}} \) from our data.](image)

![Fig. 5. \( T_1^{\text{total}} \) for different \( \Delta B \).](image)
FIG. 6. $T_1^{\text{total}}$ for different $\tau_c$. 

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