# ConceptGraphene

## New Electronics Concept: Wafer-Scale Epitaxial Graphene

## Small or medium-scale focused research project

## WP4 Spin transport devices

# Deliverable 4.2 “Comparison of spin-valves of graphene prepared with different techniques including graphene on SiC”

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## Deliverable Summary

In this report we summarize a comparative study on spin transport in several graphene types. We compare spin transport properties of epitaxial graphene on the silicon face of silicon carbide (SiC(0001)) with exfoliated graphene on silicon oxide ($SiO_2$) and on hexagonal boron nitride (h-BN). We also show preliminary spin transport measurements on epitaxial graphene on the carbon face of the crystal (SiC(000-1)) and on quasi-free-standing monolayer graphene (QFMLG), by means of hydrogen (H) intercalation.

We explain enhanced spin relaxation times and reduced spin diffusion constants in epitaxial graphene on SiC(0001) with a model for spin transport through a diffusive channel with coupled localized states. By comparison with epitaxial graphene on SiC(000-1) and QFMLG, which both do not have a buffer layer, we have found a strong indication that these localized states are present in the buffer layer of epitaxial graphene on SiC(0001).

The experimental research on spin transport in epitaxial graphene described in this report has been performed by the Groningen node of the consortium. The epitaxial graphene samples have been provided by the Linköping and Erlangen nodes.

# 1. **Introduction**

Since 2007 the Groningen group has successfully pioneered experimental work on spin transport in exfoliated graphene. Graphene is a promising material for spintronic devices, such as spin transistors or spin qubits, due to its high spin relaxation lengths of ~2μm at room temperature.[1] We are interested in spin transport properties such as the spin relaxation length, the spin relaxation time and the spin diffusion coefficient. Also we study the fundamental nature of spin relaxation mechanisms. So far, influence of the commonly used $SiO_2$ substrate seems to limit charge carrier mobility and thereby presumably spin lifetimes. Changing to epitaxial graphene gives us the opportunity to study the effect of the SiC substrate and the buffer layer on spin transport, and compare these results with previous results obtained on exfoliated graphene. Furthermore it clears the way for graphene based spintronic devices and applications in the future.

In month M18 of the ConceptGraphene project we reported on deliverable D4.1 “Report on spin transport in graphene on SiC”. In this report we described successful spin injection in en transport through epitaxial graphene on SiC(0001). We refer to report D4.1 and reference [2] for a more extensive description of the fabrication process and measurement techniques.

# 2. Comparison of graphene spin-valves prepared with different techniques

In this report we will compare the results from D4.1 with the results of spin transport measurement performed on exfoliated graphene, work that has been done earlier in our group.[1][3] We will also compare with more recent work on spin transport in high mobility exfoliated graphene on a hexagonal boron nitride (h-BN) substrate. [4]
Furthermore we performed spin transport measurements on epitaxial graphene on SiC(000-1) and on quasi-free-standing monolayer graphene (QFMLG). The graphene on SiC(000-1) samples we received from the Linköping node of the consortium. The QFMLG samples were produced using H-intercalation of graphene on SiC(0001) by the Erlangen node of the consortium.

### *2.1 Methods*

To extract spin transport properties of graphene, we use the non-local geometry of figure 1. In this geometry we separate the (charge) current path from the voltage detection circuit, in order to exclude any charge-related effects. The spin current is injected through ferromagnetic contact C2 (injector). A local spin accumulation is thereby created underneath the contact, which then diffuses through the graphene transport channel in all directions. This spin current is picked up by another ferromagnetic contact C3 (detector). To avoid conductivity mismatch and to have proper spin injection, we need an aluminum oxide (AlOx) or titanium oxide (TiOx) tunnel barrier of about 0.8 nm thick. [5]

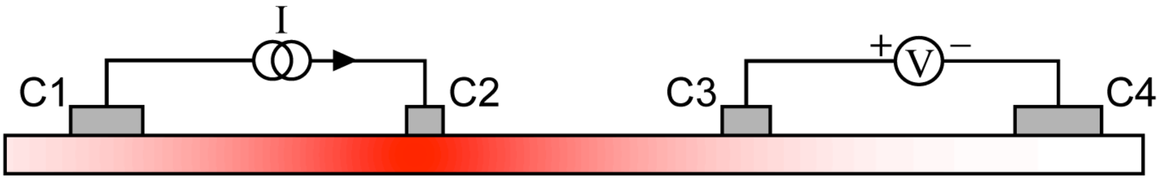


**figure 1: In the 4-terminal non-local geometry we send a spin-polarized charge current from C2 (injector) to C1, we create a spin accumulation underneath C2. This drives a pure spin current in all directions, which can be detected by measuring the non-local resistance between C3 (detector) and C4.**

If we apply an out-of-plane magnetic field, spins will start to precess during the time they spend in the channel, shown in figure 2. While sweeping the field, the non-local signal will vary, depending on how far spins on average precessed before reaching the detector (figure 4 is an example of such measurements). By fitting these so-called Hanle precession measurements with the Bloch equation describing the spin accumulation, we extract spin transport properties of the material. Thus we can determine the spin transport properties from table 1.

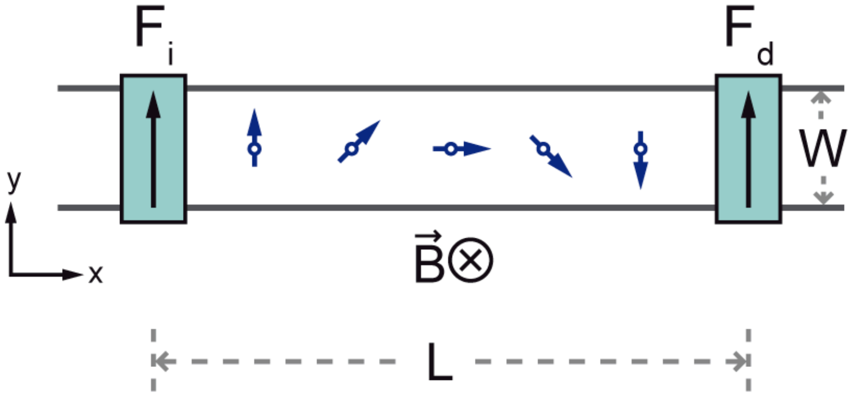


**figure 2: An out of plan magnetic field causes the injected spins to precess. Therefore the spins can arrive at the ferromagnetic detector ($F_d$) with a different orientation then they originally had when they left the injector ($F_i$).**

## *2.2 Graphene types*

Here we list brief descriptions of five different types of graphene on which we performed spin transport measurement.

1. Epitaxial graphene on SiC(0001). These results are described in more detail in D4.1. Especially interesting is the effect of the non-conducting buffer layer (see paragraph 2.5).
2. Exfoliated graphene on $SiO_2$. Reference material on which the first spin transport experiments in graphene were performed.
3. Exfoliated graphene on h-BN. Recently our group performed spin transport measurements on graphene on h-BN, which has an order of magnitude higher mobility than other graphene types.
4. Epitaxial graphene on SiC(000-1). The epitaxial graphene grown on the C-face of the SiC-crystal. It was reported to have higher mobility than Si-face grown graphene.
5. H-intercalated epitaxial graphene on SiC(0001) (quasi-free-standing, QFMLG). This material has no buffer layer between the graphene and the SiC substrate due to passivation by H-intercalation.

## *2.3 Device fabrication*

Here we address the differences between fabrication methods of exfoliated and epitaxial graphene devices and we will discuss their main advantages and disadvantages. For a more complete description of fabrication methods of epitaxial graphene spin devices we refer to the rapport on D4.1 and reference [2]. For fabrication of exfoliated graphene devices, see reference [1] and [3], for exfoliated graphene on h-BN, see reference [6].

All epitaxial graphene types allow for patterning by photolithography due to their large area. The main advantage is that we can produce several identical devices on one chip. All contact patterns down to ~1 μm are defined with an optical mask (using deep-UV photolithography), while smaller structures are defined with the use of electron beam lithography (EBL).

This fabrication process is easier and faster than preparing a sample from exfoliated graphene. We have more devices per batch, allowing for better statistics and reproducibility, given a high enough yield. Disadvantages of epitaxial graphene are the lower charge carrier mobility, higher doping and a thick insulating substrate which doesn't allow for a back-gate. Also, the device can be sensitive for local variations in the electronic and/or structural quality of the graphene. In exfoliated graphene on the other hand, we can select the best individual flakes for our devices beforehand.

Another big difference between epitaxial and exfoliated graphene is the presence of the buffer layer. This buffer layer has a large influence on spin transport through the graphene, due to the presence of localized states (see paragraph 2.5). The buffer layer can be passivated by H-intercalation, creating quasi-free-standing monolayer graphene. The buffer layer is also not present in epitaxial graphene on SiC(000-1)

### *2.4 Spin transport measurements*

We obtained data on the 5 different types of graphene described in paragraph 2.2. This data is summarized in table 1. All the experiments are performed at room temperature.

| | *Spin diffusion coefficient ($cm^2/s$)* | *Spin relaxation time (ns)* | *Spin relaxation length (μm)* |
|---|---|---|---|
| Epitaxial graphene on Sic(0001) [2] | 2.4 | 1.3 | 0.56 |
| Exfoliated graphene on $SiO_2$ [1][3] | 150-300 | 0.1-0.2 | 1-2 |
| Exfoliated graphene on h-BN [4] | 520 | 0.39 | 4.5 |
| Epitaxial graphene on Sic(000-1) [7] | 100 | 0.09 | 0.9 |
| QFMLG (by H-intercalation) on Sic(0001) [7][8] | 75 | 0.033 | 0.5 |

**table 1: Spin transport properties of different graphene types.**

Data on h-BN is from reference [4]. The high mobility clearly results in a higher diffusion coefficient and therefore in a long spin relaxation length. The higher relaxation time is due to the fact that the tunnel barriers are made of $TiO_2$ and therefore have a reduced roughness. Data on SiC(000-1) is preliminary and should still be confirmed with a second sample. Also, the results on QFMLG should be reproduced, because these measurements where performed on samples with relative low charge carrier mobility (~300 $cm^2/Vs$), while it is reported that higher carrier mobilities can be achieved in QFMLG.. We expect that the diffusion coefficient, and therefore the spin relaxation length, can be enhanced if we fabricate such a device.

As soon as the data on spin transport in all of the mentioned epitaxial graphene types is collected and summarized, we will proceed to the publication of these results. [7]

### *2.5 Modelling*

In D4.1 it was reported that we found enhanced spin relaxation times and reduced spin diffusion coefficients for epitaxial graphene on SiC(0001) (see also table 1). Furthermore, from charge transport measurements we did not obtain this reduced diffusion coefficient.

To explain these results we developed a model for spin transport in a diffusive channel with coupled localized states. Within this model (shown in figure 3) we take into account that the spin dynamics in the localized states influence the effective g-factor of the system, which will translate into an effective Bloch equation. If we analyze the measurements along this way, the results turns out to be consistent with the measured higher spin relaxation time and reduced diffusion coefficient from reference [1].

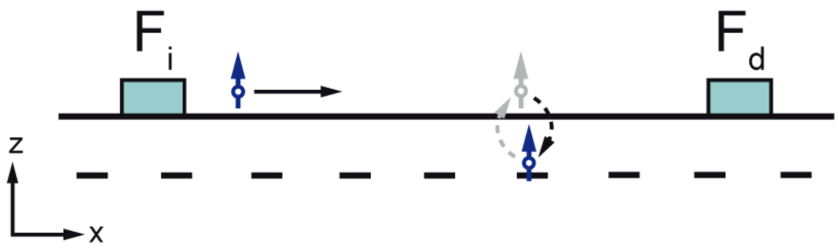
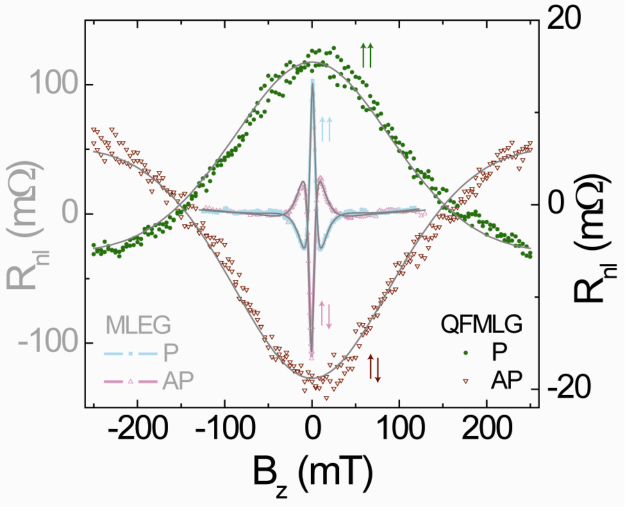


**figure 3: Extension of the Hanle geometry with localized states that are coupled to the diffusive channel. The spins can hop into these states and back into the channel while the states are not coupled with each other**

We can see the effect of the localized states if we compare Hanle precession measurement in epitaxial graphene on SiC(0001) and QFMLG (fig. 4). In QFMLG, where the buffer layer is not present due to H-intercalation, we do not observe an enhanced spin relaxation time, reduced spin diffusion time, nor a difference between diffusion coefficients obtained from charge and spin transport measurements. These different properties result in a much broader curve, as can be clearly seen in figure 4.

We submitted our results for publication to Physical Review Letters. We refer to the preprint [8] for further details concerning the localized states model.



**Comparison of Hanle spin precession in epitaxial graphene on SiC(0001) (narrow, light colored curves, scale on the left) and in QFMLG (broad, dark colored curves, scale on the right). The figure shows measurements in parallel (↑↑) and anti-parallel (↑↓) configuration.**
**The fits to the solutions of the Bloch equation are plotted in gray.**

## 3. Milestones

We achieved milestone MS6 “Spin-valve device made of graphene on SiC”. Concerning MS3 “Selection of optimal device fabrication methods”, I refer to the fabrication method described in report D4.1and reference [2].

## 4. Conclusions and outlook

In conclusion, we compared room temperature spin transport properties of several types of graphene. We can explain the enhanced spin relaxation times and reduced spin diffusion coefficients that we measured on epitaxial graphene on SiC(0001) with a spin transport model that includes localized states that are coupled to the transport channel. Preliminary data on graphene on SiC(000-1) and on QFMLG show similar spin transport properties as exfoliated graphene on $SiO_2$. This is a clear indication that the localized states are present in the buffer layer between the graphene and the SiC substrate.

We submitted our results on the localized states model for publication to Physical Review Letters. We plan to publish a comparative study of spin transport in different epitaxial graphene types, as soon as we have reproduced the preliminary data presented in this report. This study will also include temperature dependent measurements, which can give us insight in the nature of the interaction with the localized states. The next step will be the fabrication of top-gated devices for spin transport in epitaxial graphene FETs.

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