Supplementary information for 'Proximity induced room-temperature ferromagnetism in graphene probed with spin currents'

J.C. Leutenantsmeyer,* A.A. Kaverzin,* M. Wojtaszek, and B.J. van Wees

Physics of Nanodevices, Zernike Institute for Advanced Materials,
University of Groningen, 9747 AG Groningen, The Netherlands

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* These authors contributed equally to this work.
I. CHARACTERISATION OF THE YIG/GGG FILMS

FIG. 1. Magnetisation response of the 210 nm (111) YIG film on applied field and the surface topography. Left: the YIG film magnetisation in an applied perpendicular magnetic field at 293 K (black) and 70 K (red). In accordance with an easy-plane magnetisation anisotropy model it can be described as $M_z = |M| \cdot \frac{B_{app}}{B_s}$ below $B_s$ (see main text for the notations). The magnetisation $M_z$ saturates at around $B_s \sim 180$ mT at room temperature and at 250 mT at 75 K. The inset shows the in-plane magnetisation measured at 293 K showing an in-plane coercive field of less than 0.1 mT. Right: The AFM image shows a YIG substrate cleaned before the graphene transfer as described in the methods section, the scale bar corresponds to 500 nm. The topography has an RMS surface roughness of the order of 0.1 nm over a 1.75 µm by 1.75 µm square. Smaller scale corrugations (~20 nm in lateral dimension) are due to the line scanning of the image. The lateral resolution of the AFM scan is ~10 nm.

II. CHARGE AND SPIN TRANSPORT PROPERTIES OF THE GRAPHENE FLAKE

The thickness of the graphene flake is determined after exfoliation on 300 nm SiO$_2$ on a Si substrate by optical contrast (on average around 6% per single layer in our system). The discussed flake has a contrast of 5.5% from which we conclude single layer thickness. To estimate the carrier concentration we use the Shubnikov-de Haas oscillations of the longitudinal resistance at 2.2 K (Fig. 2). The reciprocal magnetic field values of the minima are plotted as a function of the peak index and shown in the inset of Fig. 2. We use only the three highest field minima to assure an accurate estimation of the carrier density. The 1/B slope of $(0.0314 \pm 0.0004)$ T$^{-1}$ corresponds to a carrier density of $n = (3 \pm 0.05) \cdot 10^{12}$ cm$^{-2}$. This value is in good agreement with our other similarly fabricated samples that show holes as carriers with densities of $n \sim 10^{12}$ to $10^{13}$ cm$^{-2}$. 
FIG. 2. The longitudinal resistance of region 1 in high fields at $T = 2.2$ K. We observe three minima of the Shubnikov-de Haas oscillations. The inset contains the reciprocal field position of the three highest minima as a function of the peak index.

Using the resistance of region 1, we deduced a carrier mobility of $(720 \pm 6.5) \text{ cm}^2/\text{Vs}$, also a typical value for our other graphene/YIG devices. With the obtained carrier density, we calculate a charge diffusion coefficient of $D_c = (66 \pm 3) \text{ cm}^2/\text{s}$.

FIG. 3. Left: The distance $d$ dependent spin signal is shown and used to calculate the spin relaxation length $\lambda$. Right: Schematic image of the graphene flake and patterned contacts. The contacts and pairs of injector-detector with different $d$ used for measurements are labelled and axes defined.

From the distance dependent spin signal (Fig. 3, left) we obtain a spin relaxation length $\lambda = (490 \pm 40) \text{ nm}$, which is in agreement with our previous samples of graphene/YIG heterostructures ($\lambda \sim 700 \text{ nm}$).
III. CHARACTERISATION OF THE GRAPHENE FLAKE AND COBALT ELECTRODES

Table I gives an overview of the measured and derived sample parameters.

| Region | $R_i$ (kΩ) | $R_d$ (kΩ) | $R_{\text{eff}}$ (kΩ) | $l \times w$ (µm$^2$) | $R$ (kΩ) | $R_{\text{sq}}$ (kΩ) | $r$ (µm) | $r/\lambda$ |
|--------|------------|------------|------------------------|------------------------|----------|-----------------------|--------|-----------|
| 1      | 1.9        | 0.89       | 1.212                  | 1.2 × 1.1              | 6.1      | 5.6                   | 0.238  | 0.49      |
| 2      | 1.9        | 0.92       | 1.240                  | 2.1 × 1.3              | 12.8     | 7.9                   | 0.203  | 0.42      |
| 3      | 0.98       | 0.92       | 0.905                  | 0.9 × 1.4              | 6.6      | 10.3                  | 0.123  | 0.25      |
| 4      | 0.98       | 0.65       | 0.751                  | 2.9 × 1.4              | 15.2     | 7.4                   | 0.143  | 0.29      |
| 5      | 0.92       | 0.65       | 0.762                  | 2.0 × 1.6              | 8.5      | 6.5                   | 0.188  | 0.38      |
| 6      | 1.90       | 0.65       | 0.969                  | 4.2 × 1.4              | 21       | 7.0                   | 0.194  | 0.40      |

TABLE I. Measured and derived parameters of the graphene flake and contacts. The table gives an overview of injector and detector resistances $R_i$ and $R_d$ measured in a 3-terminal geometry, effective contact resistance $R_{\text{eff}} = 2/(1/R_i + 1/R_d)$, length and width of the flake region $l$ and $w$, resistance $R$ of the flake region measured in 4-probe, calculated graphene square resistance $R_{\text{sq}} = R \cdot w/l$ and conductivity mismatch parameter $r = R_{\text{eff}} / R_{\text{sq}} \cdot w$ with ratio $r/\lambda$.

To analyse the influence of the contacts on the spin relaxation length $\lambda$, we calculate the conductivity mismatch parameter $r$ for the different regions. [1, 2] In the case when the contacts have the biggest contribution in spin relaxation, when $r/\lambda = 0.25$, the intrinsic spin relaxation length can be higher than the extracted value by $\sim 20\%$.

IV. DISCUSSION OF THE FULL DATA SETS. EFFECT OF THE COBALT STRAY FIELD ON THE YIG MAGNETISATION DIRECTION

We measured the spin precession at 4.7 K, 75 K and 300 K and extracted the spin signal by subtraction of the antiparallel from the parallel Hanle curve. The amplitude of the spin signal is observed to increase with decreasing temperature, which is also consistently seen in the spin valve measurements. The characteristic kink at the saturation field of the YIG magnetisation as well as the relatively linear shape of the Hanle below saturation field and the remaining spin signal up to about 600 mT are present at all temperatures. We find the kink to shift from 180 mT at room temperature to higher fields with decreasing temperature, which is in agreement with the SQUID measurements of the YIG films (see section I). To extract the spin dependent signal, we subtract the Hanle curves measured in parallel and antiparallel configurations as plotted in Fig. 4. The + designates the difference between parallel up-up and antiparallel up-down alignment of the injector/detector electrodes. The - denotes the difference between parallel
down-down and antiparallel down-up alignment. Alignment up indicates that the contact magnetisation is aligned along the y-axis (Fig. 3), in the positive magnetic field direction. Alignment down indicates the opposite, negative field, direction along the y-axis. All four curves (including trace and retrace curves for alignments + and −) show the transition point where the magnetisation of the YIG film saturates. The switches of the spin signal close to zero applied field are due to the fact that the YIG magnetisation around $B_{\text{app}} = 0$ cannot be well controlled, leading to an abrupt change of the in-plane magnetisation direction and resulting in a change of the exchange field acting on the spins.

FIG. 4. Magnetic field dependence of the spin signal obtained from Hanle precession data sets at different temperatures. Arrows indicate the YIG saturation fields.

This can be explained with the stray field arising from the contact magnetisation. The coercive field of the YIG magnetisation in the film plane (easy-plane) is smaller than 0.1 mT (see section I). Thus, even a rather small stray field can locally influence $M$. From the given geometry of the contacts we conclude that the strongest stray field is expected at the ends of the electrodes, which is typically more than 1 µm away from the graphene channel. Therefore, the
direct contribution from the stray field of the contacts to the field acting on graphene is negligible, which we confirmed with finite element modelling in COMSOL multiphysics. However, at small applied fields the YIG magnetisation can be influenced by the contact alignment which, thus, determines the exact switching behaviour of $\mathbf{M}$. Moreover, it is seen that the switching of Trace $+$ and Retrace $-$ or Trace $-$ and Retrace $+$ is symmetric with respect to the zero field, which can be understood by taking the magnetisation direction of the contacts into account.

V. POTENTIAL STRAY FIELDS ARISING FROM THE YIG FILM AND THEIR ABSENCE IN OUR MEASUREMENTS

As discussed in section IV, the stray fields arising from the contact magnetisation are expected to be negligible close to the graphene flake and cannot directly affect the spin transport in the channel. Another possible source of stray fields is the YIG film in direct vicinity to the graphene. Assuming a perfect flatness of the 210 nm YIG film and a typical size of $5 \times 5$ mm, no stray fields are expected. However, it was shown in by Dash et al. [3] that the finite roughness of the surface of the magnetic material can lead to non homogeneous in-plane and out-of-plane stray fields. Under the assumption of a perfectly flat graphene flake, the out-of-plane stray fields average out spatially, as the total magnetic flux through the graphene surface has to be zero. With finite roughness of the graphene both in-plane and out-of-plane fields can have a non-zero average value.

In our case the roughness of the YIG film is $\sim 0.1$ nm (section I), therefore, we expect a negligible magnitude of any YIG stray field. Moreover, based on our analysis we can exclude the effect of stray fields because of the following reasons. Firstly, to explain our results with stray fields only, a magnitude of the order of 0.2 T, comparable to the YIG saturation field, would be required. Such a large average stray field cannot originate from the measured YIG roughness. Secondly, the fitting of both out-of-plane Hanle measurements (Fig. 3, main text) and in-plane rotation measurement (Fig. 4, main text) leads to a similar magnitude of the exchange field. Thus, the effect cannot be explained by stray fields as they are expected to be very different for in-plane or out-of-plane magnetisation configurations.

Furthermore, we exclude the effect of stray fields using measurements of the Hall effect in similarly fabricated samples of graphene on YIG with non-magnetic titanium/gold contacts. Similarly to Ref. [4], we measure the transverse resistance of the graphene flake in a perpendicular applied magnetic field. After antisymmetrisation of the data the linear component is determined by a linear fit at high fields (between 400 and 650 mT, hatched area of Fig. 5) and subtracted. The remaining non-linear component is shown in Fig. 5. If present, the stray field is expected to saturate along with the perpendicular saturation of YIG magnetisation and, therefore, should be seen in the non-
FIG. 5. **Characterisation of the Hall effect on graphene on YIG.** The transverse resistance ($R_{xy}$) of a graphene flake is measured and antisymmetrised (inset). After subtraction of the linear Hall effect (45.28 Ω/T, measured in the hatched area of the inset) non-linear component remains, which has a magnitude of roughly 0.3 Ω at $T = 75$ K.

The observed non-linear contribution is $\sim 100$ times smaller than the linear component. This implies that the maximal strength of the stray field has to be 100 times smaller than the applied magnetic field ($\sim 0.2$ T), i.e. $\sim 0.002$ T. This is also 100 times lower than the extracted strength of the exchange field and provides an experimental evidence to exclude stray fields as a possible explanation of our results.

**VI. ABSENCE OF A PARALLEL MAGNON TRANSPORT CHANNEL**

As shown by Cornelissen et al. [5] and Goennenwein et al. [6] a spin accumulation induced in a material in proximity with YIG can excite a magnon current in the FMI, leading to a parallel spin transport channel from the injector to the detector. We can exclude the existence of an additional spin transport channel in our graphene/YIG system, since the magnon transport process is suppressed at low temperatures [6, 7], while we find an increase of the spin signal, confirming that the signal is not carried by magnons.
VII. COMPARISON TO A REFERENCE SAMPLE

FIG. 6. Comparison to a reference sample. The spin precession data of the additional reference sample (red squares, 1 µm contact spacing, 70K) is plotted together with the same measurement for main text sample (black line, 1.2 µm contact spacing, 75 K). Despite similar fabrication steps, we find a higher contact impedance and an increased noise level in the reference sample. However, we are able to observe a comparable magnitude of the spin signal as well as the characteristic features like the relatively linear shape at lower fields and the kink at the perpendicular saturation field of the substrate. The inset contains a non-local spin valve measurement of the reference sample.

VIII. FULL SET OF SPIN TRANSPORT MODULATION DATA

FIG. 7. Spin modulation in region 1 at room temperature and 75 K. The spin signal in region 1 (1.2 µm) at 75 K is modulated by 57%. The modulation increases at room temperature to 77% (100 mΩ).
IX. COMPARISON WITH SPIN TRANSPORT MODULATION IN METAL/YIG SYSTEMS

Recent reports from Villamor et al. [8] and our group [9] demonstrated the modulation of spin currents in Cu and Al films on YIG substrates. It was found that the spin signals can be modulated by an in-plane rotation of the YIG magnetisation. Although the interaction in this case is also induced by an exchange interaction, in metals the effect causes predominantly an absorption of spins perpendicular to the YIG magnetisation.

This description arises from circuit theory which is applicable to diffusive metal systems, which support many disordered conduction channels. In case of graphene, this concept does not apply since graphene is a strictly two dimensional electronic system. The effect of the YIG on the spins in the graphene channel is not caused by absorption but by induced precession, which arises from the exchange interaction, which causes a spin-splitting in the band structure. Our measurements clearly indicate the presence of this exchange interaction since we can extract the strength from our data. It implies that the electronic states of the graphene are modified and we can call the
graphene ferromagnetic. Note that in a metal system the density of states close to the interface could in principle
also become spin polarised. However, it would be restricted to the first atomic layer(s), whereas the experiments in
metals are done in much thicker layers, where circuit theory is applicable.

X. MODELLING

To model the observed data we solve the one dimensional Bloch diffusion equation for a total effective field acting
on the spins:

\[ 0 = D_s \nabla \mu_s - \frac{\mu_s}{\tau_s} + \frac{g \mu_B}{\hbar} \left( \mathbf{B}_{\text{app}} + \mathbf{B}_{\text{exch}} \right) \times \mathbf{\mu}_s \]  

(1)

where \( \mathbf{\mu}_s(x) = (\mu_x(x), \mu_y(x), \mu_z(x)) \) and the magnetic field \( \mathbf{B}_{\text{tot}} \) is the vector sum of the external applied magnetic
field and exchange field (\( \mathbf{B}_{\text{tot}} = \mathbf{B}_{\text{app}} + \mathbf{B}_{\text{exch}} \)). The equation can be solved with the boundary condition for the
spin accumulation \( \mathbf{\mu}_s(x) = (0, 0, 0) \) at \( x = \pm \infty \) and the assumption that the spins are injected only in y-direction,

\[ \frac{\partial}{\partial x} \mathbf{\mu}_s(x) \sim (0, \mu_y, 0) \text{ at } x = 0. \]

The analytical solution for the y-component of the spin accumulation reads:

\[ \mu_y(x) = A \left[ \frac{B_y^2}{B^2} e^{-\frac{x}{\lambda}} + \frac{B_y^2 (B_z - B_x)^2}{2B^2(\alpha_1^2 + \alpha_2^2)} (B_z B_x - B_y B_y - B_z B_y + B_z B_y)^2 \right] \left( \alpha_1 \cos \left( \frac{x}{\lambda} \right) - \alpha_2 \sin \left( \frac{x}{\lambda} \right) \right) e^{-\alpha_1 \frac{x}{\lambda}}, \]  

(2)

where \( A \) is a scaling parameter, \( \alpha_{1(2)} = \frac{1}{\sqrt{2}} \sqrt{1 + \sqrt{1 + (\omega \tau_s)^2}}, \omega = \frac{g \mu_B}{\hbar} B = \frac{g \mu_B}{\hbar} |\mathbf{B}_{\text{tot}}| \) and \( \tau_s = \lambda^2 / D_s \).

For relevant cases when either \( B_x = 0 \) or \( B_y = 0 \) the expression for \( \mu_y(x) \) can be simplified:

\[ \frac{B_y^2 (B_z - B_x)^2 + (B_z B_x - B_y B_y - B_z B_y + B_z B_y)^2}{2B^2(B_x B_y - B_y B_z - B_z B_x)} \cdot \left\{ \begin{array}{ll} \left( \frac{B_y}{B} \right)^2, & \text{when } B_x = 0; \\ 1, & \text{when } B_y = 0. \end{array} \right. \]  

(3)

The obtained expression Eq. 2 is used to fit three types measurements. First, we fit the Hanle precession data when
the external field is applied perpendicular to the sample plane (Fig. 3c and 5a from the main text). \( A, \lambda, \tau_s \) and
\( |\mathbf{B}_{\text{exch}}| \) are used as parameters. The best fit is obtained with \( |\mathbf{B}_{\text{exch}}| \sim 0.2 \text{ T}, \tau_s \sim 27 \text{ ps} \) and \( \lambda \sim 1.8 \text{ \mu m} \), Fig. 9 left.

From the modelling we conclude that when \( \mathbf{M} \) is aligned with \( \mathbf{B}_{\text{app}}, \mathbf{B}_{\text{app}} \) and \( \mathbf{B}_{\text{exch}} \) have the same sign.

Secondly, we fit the relative modulation of the spin signal as a function of the distance between electrodes when
the magnetisation of the YIG is rotated in the sample plane (Fig. 4, main text) with \( A, \lambda \) and \( \tau_s \cdot |\mathbf{B}_{\text{exch}}| \). The best
fit is obtained with \( \lambda \sim 1.8 \text{ \mu m} \) and \( \tau_s \cdot |\mathbf{B}_{\text{exch}}| = 7.8 \text{ ps-T}, |\mathbf{B}_{\text{exch}}| \sim 0.2 \text{ T} \) and \( \tau_s \sim 39 \text{ ps} \), Fig. 9 right.

Lastly, we fit the in-plane Hanle precession when the direction of applied field is fixed along the x-axis, within
the sample plane but perpendicular to the easy-axis of the contacts magnetisation (Fig. 10 left). Due to smaller
in-plane shape anisotropy compared to out-of-plane, we take the rotation of the contact magnetisation in-plane into
FIG. 9. Influence of the spin relaxation length and exchange field on the Hanle spin precession and spin signal modulation. The derived model is used to fit experimental results. The circles represent the experimental results and solid lines the model curves for different parameters. Left: fitting of the Hanle dependence from Fig. 3 (main text). Right: fitting of the relative modulation dependence derived from Fig. 4 (main text).

account. While the out-of-plane saturation field is around 1.2 T, the in-plane saturation field along the x-axis is between 100 – 200 mT, leading to a deviation of the direction of the injected spins relative to the y-axis. The contact magnetisation and, therefore, the spin accumulation is calculated with an easy axis magnetic anisotropy model using the saturation field of the contact magnetisation as an additional parameter. With the previously obtained values $\lambda \sim 1.8 \mu$m and $|B_{\text{exch}}| \sim 0.2$ T we can qualitatively fit the measured dependencies with $\tau_s \sim 15$ ps and a contact saturation field of 140 mT. It implies that above 140 mT both contact magnetisations and the YIG magnetisation are aligned with the external magnetic field, leading to a maximum spin signal. However, we find a further increase of the spin signal of unknown origin when applying magnetic fields up to 7 T (Fig. 10 right).
FIG. 10. **In-plane Hanle spin precession.** Left: Hanle curves with the magnetic field applied in-plane along x-axis for parallel (black squares) and antiparallel (red squares) alignments fitted with the model (solid lines) taking into account the in-plane rotation of the contact magnetisation with the applied field. Right: Measurement of the in-plane Hanle curve up to 7 T.

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