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Spin Transport in High-Quality Suspended Graphene Devices

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S Supporting Information

ABSTRACT: We measure spin transport in high mobility suspended graphene ($\mu \approx 10^5 \text{ cm}^2/(\text{V s})$), obtaining a (spin) diffusion coefficient of $0.1 \text{ m}^2/\text{s}$ and giving a lower bound on the spin relaxation time ($\tau_s \approx 150 \text{ ps}$) and spin relaxation length ($\lambda_s = 4.7 \text{ }\mu\text{m}$) for intrinsic graphene. We develop a theoretical model considering the different graphene regions of our devices that explains our experimental data.

KEYWORDS: Suspended graphene, spin transport, Hanle precession

The prospectives for graphene spintronics are very positive with theoretical predictions of spin relaxation times (τ_s) in the order of hundreds of nanoseconds and higher^{1,2} due to the lack of nuclear spin for carbon's most common isotope (^{12}C) and weak intrinsic spin–orbit coupling. On the other hand, the experimental results typically find τ_s of hundreds of picoseconds.^{3–5} In order to clarify the limitations and mechanisms for spin relaxation in monolayer graphene devices a lot of effort has been done by both experimentalists^{3–9} and theoreticians^{2,10–12} but up to now this topic is still under debate. Theoretical results pointed out that the D'Yakonov–Perel mechanism¹³ and locally enhanced spin–orbit coupling due to impurities and lattice deformations^{2,10} are the most probable mechanisms for spin relaxation for measurements in graphene spintronic devices.^{2,11,12} Since all devices studied so far were fabricated on substrates (SiO_2 ^{3–8} or SiC ¹⁴) in which the intrinsic properties of graphene are masked due to the electronic coupling to the substrate,¹⁵ no previous study was able to confirm these recent theoretical predictions. In order to address this problem, we study spin transport in high-mobility suspended graphene devices. By removing the substrate, thereby suspending the graphene flake, we are not only capable of achieving a high quality device with low contamination,^{16,17} but we can also investigate how the absence of the rough SiO_2 substrate influences the spin transport in graphene. Moreover it opens the possibility to exploit the exquisite mechanical properties of graphene¹⁸ and to study how pseudomagnetic fields and strain^{19,20} affect spins in graphene, paving the way for graphene in the field of spin-nanomechanical applications.^{21–23}

The effective spin–orbit (SO) field that the spins experience in graphene is directly related to the density of impurities and adatoms.^{10,12} This effective SO field causes a momentum-dependent spin precession, resulting in spin decoherence and relaxation. This mechanism is known as the D'Yakonov–Perel mechanism for spin relaxation.¹³ Assuming such a mechanism

for spin relaxation²⁴ with an effective SO coupling Δ_{SO} , the spin relaxation time behaves like^{12,13}

$$\frac{1}{\tau_s} = \frac{4\Delta_{\text{SO}}^2}{\hbar^2} \tau_p \quad (1)$$

where τ_p is the momentum relaxation time and $\hbar$ is the reduced Planck constant. With the reduction of adatoms and impurities, we should obtain a low value of Δ_{SO} approaching the theoretical predictions^{2,25,26} ($\Delta_{\text{SO}}^{\text{theory}} \approx 10 \text{ }\mu\text{eV}$) and a high value for the momentum relaxation time $\tau_p > 0.1 \text{ ps}$. Since τ_s increases quadratically with the reduction in Δ_{SO} , we expect that for clean samples we reach the initial theoretical limits of $\tau_s > 10 \text{ ns}$.

At a first glance, suspending the graphene flake to obtain a very high quality device seems the best approach. But the main challenge is that the most common technique to suspend graphene flakes^{16,17} is acid-based and therefore not compatible with ferromagnetic metals necessary for spin transport. The reason is that the acid used to etch the SiO_2 underneath the graphene also etches away the ferromagnetic metals used for the electrodes. Also, devices produced by the standard technique are typically short and are in the order of $1 \text{ }\mu\text{m}$ or less to avoid that the graphene flakes collapse when a gate-voltage is applied. This is undesirable for spin precession measurements since the time for one period of precession for low magnetic fields ($B < 1 \text{ T}$) have to be in the same order as the diffusion time of the spins in order to obtain a Hanle precession curve²⁷ showing a complete spin precession. To overcome these issues, we developed a polymer-based method²⁸ in which we are able to produce flakes suspended over long distances and contacted by ferromagnetic electrodes

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Figure 1. (a) The graphene flakes are exfoliated on a LOR film spin-coated on a Si/SiO₂ substrate followed by the deposition of Co contacts with aluminum oxide insulating barriers to avoid the conductivity mismatch. (b) Local 4-probe measurement geometry to characterize the graphene resistance as a function of the gate-voltage applied to the highly doped Si substrate. A second electron beam lithography step is performed to suspend the central region. (c) Scheme of the current annealing setup used to clean the graphene devices. The high DC bias is applied to the outer electrodes to avoid the degradation of the inner contacts since these are to be used for spin injection/detection. (d) Scanning electron micrographs of a typical device. The scale bars are 1 μm . (e) Local 4-probe resistance (top) and mobility (bottom) versus carrier density and gate-voltage after the current annealing procedure at 4.2 K. The carrier density shown corresponds to the carrier density in the central suspended region (see main text).

with highly resistive barriers. In our process, the graphene flakes are exfoliated on top of a 1 μm thick lift-off resist (LOR) film spin-coated on a Si/SiO₂ (500 nm) substrate. Single layer graphene flakes to be used in our devices are selected by optical contrast.^{29,30} The highly doped Si substrate is used to apply a back-gate voltage V_g that induces a charge carrier density according to $n = \alpha(V_g - V_0)$, where $\alpha = 0.45 \times 10^{10} \text{ cm}^{-2} \text{ V}^{-1}$ is the effective gate capacitance and V_0 is the position in gate voltage of the minimum of conductance. Using standard electron beam lithography (EBL) and metal evaporation methods we deposit the 1 nm thick Al₂O₃ tunnel barriers and the 60 nm Co contacts (Figure 1a). A second EBL step is used to make parts of the graphene flake suspended (Figure 1b). The detailed device fabrication is described in the Supporting Information. The resistance of our contacts (usually $R_C \sim 20 \text{ k}\Omega$) and the gate-voltage dependence of our devices are characterized at the beginning of every set of measurements. Before the quality improvement via the current annealing procedure (described in the next paragraph), the devices typically show high p-doping and only a small change of the resistance as a function of V_g is observed.^{28,31}

To improve the quality of our suspended graphene devices we perform current annealing³² in vacuum (at a pressure better than 10^{-6} mbar) at a base temperature of 4.2 K (see Supporting Information). Since the contact resistance of the spin injector and detector in our samples has to be kept at high values ($>10 \text{ k}\Omega$) to avoid the impedance mismatch and contact induced spin relaxation,^{4,7,33} we use the outer electrodes to apply the high current densities capable of heating the graphene flake to high temperatures (Figure 1c). As shown in Figure 1b,c, our devices are composed of two supported outer parts with a central suspended part. During the annealing the graphene flake is cooled by the contacts and the substrate in the supported parts and mainly heats up in the suspended region. When a

sufficiently high temperature is achieved, the polymer residues and contaminants are removed from the suspended central region. Finally the high quality of the devices can be verified via the behavior of the square resistance of the central region (R_{sq}) as a function of the gate-voltage (Figure 1e). It is worth noting that the value and the gate dependence of the sheet resistance of the supported regions do not change significantly after the current annealing procedure. This means that the values of carrier density presented here are representative for the central suspended region. We obtained two high quality devices in different flakes with mobilities higher than $\mu = 10^5 \text{ cm}^2/(\text{V s})$. One of the devices was current annealed twice and showed a mobility of $\mu \approx 1.2 \times 10^5 \text{ cm}^2/(\text{V s})$ after the first current annealing and $\mu \approx 3 \times 10^5 \text{ cm}^2/(\text{V s})$ after the second. This device was also characterized for charge and spin transport in between the two current annealing steps. Here we show the results for this representative high mobility sample after the second current annealing. The separation between the inner electrodes (suspended region) is $L = 2.5 \mu\text{m}$ and the electronic mobility is $\mu \approx 3 \times 10^5 \text{ cm}^2/(\text{V s})$ (Figure 1e). Similar results for both charge and spin transport were obtained in the other devices.

The values for mobility were obtained in two different ways: either by using the equation $\mu = \sigma/(ne)$ and taking the value of μ at a carrier density of $n = 1 \times 10^{10} \text{ cm}^{-2}$, or by fitting the conductivity curves with the formula $1/\sigma = 1/(ne\mu + \sigma_0) + \rho_s$,^{34,35} where σ is the graphene conductivity, e is the elementary charge, σ_0 is the conductivity at the charge neutrality point and ρ_s is the contribution of short-range scattering. The values obtained by both ways are consistent with each other and are in the order of $10^5 \text{ cm}^2/(\text{V s})$. We often observe a background resistance in our $R_{sq}(V_g)$ curves (Figure 1e). The most probable source for this background resistance is the noncleaned region underneath and/or close by the contacts. We did not subtract

Figure 2. (a) The 4-probe nonlocal device configuration for Hanle precession and spin-valve measurements. For the precession measurements the two inner contacts are aligned in a parallel (antiparallel) configuration and the nonlocal resistance is measured as a function of a perpendicular magnetic field $B_{\perp}$. (b) A measured Hanle precession curve (black circles) with a fit using the solutions for the Bloch equations (red line). The spin signal R_s is recorded as a function of the perpendicular magnetic field. Left inset: the nonlocal resistance as a function of the parallel magnetic field. The positive (negative) values of R_{nl} are due to parallel (antiparallel) alignment of the inner electrodes. Right inset: the nonlocal resistance as a function of the perpendicular magnetic field for the parallel (black) and antiparallel (red) configuration of the contacts. (c) Spin and charge diffusion coefficients D_s (red squares) and D_c (dashed line) respectively, spin relaxation time τ_s and spin relaxation length λ_s as a function of the carrier density in the central suspended region. All the measurements shown were performed at 4.2 K.

this background resistance in our calculations since this would only lead to higher mobility values.

To perform the spin-transport measurements we use a nonlocal geometry where the charge current path is separated from the voltage contacts to exclude spurious signals⁶ (Figure 2a). When we sweep the in-plane magnetic field we can align the two inner ferromagnetic contacts in a parallel or antiparallel configuration. To extract the spin diffusion coefficient D_s and spin relaxation time τ_s , we perform Hanle precession measurements and use the solutions for the Bloch equations in the diffusive regime.^{7,27} The Hanle precession measurements are done by measuring the nonlocal resistance as a function of an applied perpendicular magnetic field. To eliminate background signals in our analysis we fit the data for the total spin-signal given by $R_s = (R_{nl}^{\uparrow\uparrow} - R_{nl}^{\uparrow\downarrow})/2$, where $R_{nl}^{\uparrow\uparrow(\downarrow\downarrow)}$ is the nonlocal resistance in the parallel (antiparallel) configuration of the inner electrodes (Figure 2b). The values for D_s and τ_s obtained for the suspended nonannealed samples are typically $D_s \approx 0.02 \text{ m}^2/\text{s}$ and $\tau_s \approx 200 \text{ ps}$, giving a spin relaxation length of $\lambda_s = (D_s \tau_s)^{1/2} \approx 2 \mu\text{m}$, showing no dependence on the temperature (from room temperature down to 4.2 K). Since the results for the nonannealed samples show no substantial difference from measurements done in fully LOR supported devices (see Supporting Information), we can conclude that the substrate is not the main factor limiting the spin relaxation for our samples before the cleaning procedure. When we consider roughness effects, this invariance of the spin relaxation time in our measurements without the rough substrate is in agreement with the results by Avsar et al.,⁵ where it is shown that ripples in the graphene flakes have minor (or no) effects on the spin transport parameters. Also, the widths of our contacts in our samples are much smaller than the spin relaxation length. So we

believe that the regions underneath the contacts do not change significantly the spin transport properties in our measurements.

After cleaning by current annealing, we extract τ_s and D_s as a function of the carrier density via Hanle precession measurements (Figure 2c). Comparing our results for the clean high-mobility samples to previous studies on SiO_2 supported devices^{3–8} we observe an approximately 3 times higher spin diffusion coefficient while the values for the spin relaxation time remain similar. Note that the experimental conditions allow us to extract a lower bound for the spin relaxation time in the suspended graphene region, this leads to important conclusions as we will discuss below. All the spin transport measurements presented here were performed at low temperatures (4.2 K).

We start by discussing the results for the spin diffusion coefficient. In Figure 2c, we have a comparison of the values of D_s extracted from the Hanle precession measurements (red squares) with the value of the charge diffusion coefficient (dashed line) given by the Einstein relation $D_c = 1/(R_{sq} e^2 \nu(E))$, where $\nu(E)$ is the density of states of graphene at the energy E . In this case, the square resistance, R_{sq} , was extracted by local 4-probe measurements, as depicted in Figure 1b. It can be seen that D_s and D_c are in reasonable agreement, which is in accordance to previous works on exfoliated monolayer graphene.³

We now turn our attention to the results for the spin relaxation time τ_s (Figure 2c). If the spin relaxation time in graphene is limited by a locally enhanced SO coupling^{11,12} due to impurities and adatoms, we would expect a high τ_s for a high mobility sample, since an increased mobility is related to a reduction in the density of impurities. Intriguingly, in our results for high mobility graphene spin-valves, in which the mean free path is in the order of a micrometer, τ_s is still in the

order of hundreds of picoseconds. When we calculate the spin relaxation length $\lambda = (D_s \tau_s)^{1/2}$ we obtain large values, up to $\lambda = 4.7 \mu\text{m}$, even with the low spin relaxation time in our samples. But one question remains: what limits the measured spin relaxation time in our high-quality devices? To address this issue we performed numerical simulations of spin transport in our devices.

To properly represent our devices we extend the model adopted by Popinciuc et al.⁷ We consider our devices as a three-part system separated by two boundaries. The central part represents the suspended region and the two identical outer parts represent the supported regions (Figure 3a). The spins

Figure 3. (a) A cartoon of the system used in our simulations: two identical and semi-infinite parts connected by one inner region with length L , all of width W . The spins are injected at the left boundary ($x = 0$) and detected at the right boundary ($x = L$). (b) A graph showing the results obtained for the diffusion constant D_{fit} by fitting the simulated data with D_i (D_o) constant at the value represented by the dotted black line and changing D_o (D_i) following the dashed gray line in blue circles (red stars). (c) Results for the spin relaxation time τ_{fit} by fitting the simulation data with τ_i (τ_o) constant at the value represented by the dotted black line and changing τ_o (τ_i) following the dashed gray line in blue circles (red stars). The resistivities of the three regions were kept at 1 k Ω for the results in (b) and (c).

are injected at the left boundary by an injector of polarization P_{c1} and contact resistance R_{c1} and detected by a contact at the right boundary with polarization and contact resistance P_{c2} and R_{c2} respectively. The spin diffusion coefficients, spin relaxation times and the conductivities, denoted by $D_{i(o)}$, $\tau_{i(o)}$, and $\sigma_{i(o)}$, respectively, for the inner (outer) parts can be defined independently. We assume that the spin accumulation $\vec{\mu}_s(x)$ obeys the Bloch equation for diffusion in one-dimension with

an applied magnetic field $\vec{B}$: $D_s \nabla^2 \vec{\mu}_s - (\vec{\mu}_s / \tau_s) + \gamma \vec{B} \times \vec{\mu}_s = 0$, where $\gamma = g \mu_B \hbar^{-1}$ is the gyromagnetic ratio, g is the g-factor, μ_B is the Bohr magneton, and $\hbar$ is the reduced Planck constant. For the boundary conditions, we assume that $\vec{\mu}_s$ is continuous and goes to zero at $x = \pm\infty$, the spin current at the right boundary is continuous, and at the left boundary it is discontinuous by a value determined by the spin injection. We also include the contact induced spin relaxation due to back-diffusion,⁷ although for contact resistances in the order of those we encounter in our experiments, our simulations showed no substantial difference from the results considering the limit of infinite contact resistances. Details of the model and simulation are included in the Supporting Information.

By adding a perpendicular magnetic field $\vec{B} = B \hat{z}$ the spins can precess and the voltage at the detector electrode is calculated as a function of B , simulating the Hanle precession measurements. The obtained data is then fitted the same way we fit our experimental data, with the solutions to the Bloch equation for a homogeneous system, and effective values for D_s and τ_s are obtained and now denoted as D_{fit} and τ_{fit} respectively. These effective values are then compared to the values used in the simulation.

Since we have four different parameters to consider (τ_i , τ_o , D_i and D_o), we change them one by one, keeping the others at a constant value. We keep the sheet resistance of the three regions at the same value of 1 k Ω . The effect of varying the sheet resistance of the inner and outer regions are presented in the Supporting Information. Keeping $\tau_i = \tau_o = 200$ ps and $D_i = 0.1 \text{ m}^2/\text{s}$, we varied D_o from 0.01 to 0.5 m^2/s and extract D_{fit} (Figure 3b, blue circles), which we compare to D_i (dotted black line) and D_o (dashed gray line). It can be seen that a change in the spin-diffusion coefficient of the outer parts does not influence the results obtained by the Hanle precession fits in the studied range. For all values of D_o , we always obtained a value close to the value of D_i , $D_{\text{fit}} \approx 0.1 \text{ m}^2/\text{s}$. Keeping now D_o constant and varying D_i (Figure 3b, red stars) we confirm that from the Hanle precession analysis we always obtain a value D_{fit} very close to the actual value of D_i (dashed gray line). Such a result agrees with our experimental measurements, where the values obtained for D_s from the Hanle precession measurements are in good agreement with the charge diffusion coefficient, D_c , for the inner suspended part. In other words, the diffusion coefficient we measure for our inhomogeneous devices is determined by the one in the central high mobility region.

Having confirmed that the results obtained for the diffusion constant from our experiments and simulations follow the same trend, we now analyze the effect of the spin relaxation times. We fix the spin diffusion coefficients at $D_i = 0.1 \text{ m}^2/\text{s}$ and $D_o = 0.01 \text{ m}^2/\text{s}$, which are approximately the values we find in our experiments, and keep $\tau_i = 200$ ps constant (dotted black line). We then vary τ_o from 20 to 2000 ps (Figure 3c, blue circles). The values obtained from the fit of the simulated data for τ_{fit} follow the increase in τ_o (dashed gray line) and only start deviating at large values $\tau_o \approx 2000$ ps. On the other hand, when τ_o is kept constant at 200 ps and we change τ_i , the values obtained from the fits seem to be determined by the value of the spin relaxation time in the outer regions. From our calculations, this result holds for $D_o \leq D_i$, which falls in our experimental range.

The results for the spin relaxation time in our model can be explained in a qualitative manner. Because of its diffusive motion, a part of the injected spins spend some time in the

supported region before crossing the suspended region reaching the detector electrode (right boundary). For a sufficiently low diffusion coefficient in the supported part, the time spent there is enough to relax most of the spins in the case of a short spin lifetime. Considering $D_s \approx 0.1 \text{ m}^2/\text{s}$ from our experimental results, the average time the spins take to diffuse through the central suspended region is about $\tau_d = (L^2/(2D_s)) \approx 30 \text{ ps}$ for $L = 2.5 \text{ }\mu\text{m}$. Therefore we should see a decrease in the effective spin relaxation time τ_{fit} in case the average time the spins take to diffuse through the central region is in the order of the spin relaxation time of this region (τ_i), since most of the spins will relax before reaching the detector electrode. This consequence can be seen in the right graph of Figure 3c. If τ_i is longer than τ_d , the main limiting factor in the effective relaxation time is τ_o .

Now we will compare our theoretical results with the experimentally obtained values. As said before, we have a good representation of D_s for the central suspended region from the values extracted by the experimental Hanle curves. On the other hand, the obtained values for the spin relaxation time are apparently limited by the “dirty” outer regions. By performing Hanle precession measurements in fully LOR supported spin-valves, we obtain values of $\tau_s \approx 150 \text{ ps}$ and $D_s \approx 0.02 \text{ m}^2/\text{s}$, which support our conclusions. This means that the spin relaxation time extracted from our Hanle precession measurements represents a lower bound on the spin relaxation time of the high quality central region. As far as we are aware, this effect of inhomogeneity in a spin-valve was never reported and it is of importance to experiments in any inhomogeneous system and not exclusively for graphene. For systematic studies in nonlocal spin valves as a function of the local properties of the central region (e.g., mobility, conductivity or carrier density), one must consider this inhomogeneity effects in the analysis, otherwise it may lead to erroneous interpretation of the experimental data.

Although we are not able to determine the actual spin relaxation time in the high mobility region, the experimental conditions allow us to take a very important conclusion with regard to the spin relaxation mechanism. If we consider the D’Yakonov–Perel as the main mechanism for spin relaxation in graphene, we can assume that τ_s relates with the momentum relaxation time τ_p according to¹³ eq 1. Since the charge diffusion coefficient relates to τ_p by $D_c = ((v_F^2)/2)\tau_p$, we can extract for our high mobility samples $\tau_p \approx 0.2 \text{ ps}$. We can then obtain an upper bound for the average spin–orbit coupling $\Delta_{\text{SO}} \leq 50 \text{ }\mu\text{eV}$ using eq 1. It is important to notice that the value for Δ_{SO} that we estimate is an upper bound that is calculated from the experimentally determined lower bound on the spin relaxation time in the suspended high quality graphene. Applying the same procedure for typical SiO_2 supported graphene spin valves $\tau_p = 0.04 \text{ ps}$ and $\tau_s = 200 \text{ ps}$ we obtain an average SO coupling of $\Delta_{\text{SO}} = 110 \text{ }\mu\text{eV}$. If this value was intrinsic for graphene, we would obtain a strong reduction in the spin relaxation time in our high quality graphene devices down to $\tau_s = 50 \text{ ps}$. This value for τ_s is comparable to the time the spins take to diffuse through the central suspended region (τ_d) and smaller than the spin relaxation time for the nonsuspended regions. This means that if the value for SO coupling obtained in the SiO_2 devices was an intrinsic value for graphene we should be able to observe a very low τ_s , which is not the case. From this result we can conclude that the nature of the SO fields in the graphene devices observed so-far is not due to graphene’s properties, but due to extrinsic effects. The value obtained for Δ_{SO} in our suspended graphene devices is

about a factor of 2 lower than what is usually obtained in the low mobility SiO_2 supported devices, although it is still five times higher than the theoretical limit of $\Delta_{\text{SO}} \approx 10 \text{ }\mu\text{eV}$.^{2,25,26} Our results are in agreement with recent theoretical predictions that Δ_{SO} scales with the presence of adatoms,^{10,12} so higher spin lifetimes in high quality graphene flakes should be expected.

In conclusion, we performed spin transport in suspended high mobility monolayer graphene devices contacted by ferromagnetic leads. We showed that electronic mobilities above $100\,000 \text{ cm}^2/(\text{V s})$ for the suspended region can be achieved in our devices via current annealing. Our measurements showed an increase up to 1 order of magnitude in the spin diffusion coefficient when compared to SiO_2 supported devices and very large spin relaxation lengths ($\lambda = 4.7 \text{ }\mu\text{m}$) were obtained. We did not observe a change in the measured spin relaxation time when comparing our results to the traditional SiO_2 supported devices. This effect is explained by considering a simple model that takes the inhomogeneity in our devices into account, considering not only the suspended regions of the devices, but also the supported part. We observe that the spin transport measurements of our high mobility graphene devices strongly depend on how spins interact in the lower mobility graphene parts directly connected to it. In a similar way, this inhomogeneity effect is expected to strongly affect the performance of a typical graphene nanodevice when connected to other graphene nanodevices or interconnects with different spintronic properties (due to possible edge roughness or other kind of spin scattering in the system). For future works, both in graphene- and nongraphene-based devices, one must take this effect into account in order to make concise conclusions. We were also able to give a higher bound for the spin–orbit coupling in our devices of $50 \text{ }\mu\text{eV}$, which is a factor of 2 lower than of those encountered in SiO_2 supported devices.

■ ASSOCIATED CONTENT

📄 Supporting Information

The Supporting Information contains a more detailed description of the sample preparation, the current annealing procedure to achieve the high mobilities in our devices, a detailed description of the simulation methods and model, and the effect of the sheet resistance in the Hanle precession curves. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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