Omnidirectional spin-to-charge conversion in graphene/NbSe₂

Josep Inglà-Aynés*, Inge Groen, Franz Herling, Nerea Ontoso, C K Safeer, Fernando de Juan, Luis E Hueso, Marco Gobbi, C K Safeer and Félix Casanova

Abstract
The conversion of spin currents polarized in different directions into charge currents is a keystone for novel spintronic devices. Van der Waals heterostructures with tailored symmetry are a very appealing platform for such a goal. Here, by performing nonlocal spin precession experiments, we demonstrate the spin-to-charge conversion (SCC) of spins oriented in all three directions (x, y, and z). By analyzing the magnitude and temperature dependence of the signal in different configurations, we argue that the different SCC components measured are likely due to spin–orbit proximity and broken symmetry at the twisted graphene/NbSe₂ interface. Such efficient omnidirectional SCC opens the door to the use of new architectures in spintronic devices, from spin–orbit torques that can switch any magnetization to the magnetic state readout of magnetic elements pointing in any direction.

1. Introduction
Efficient spin-to-charge conversion (SCC) is a crucial ingredient for spintronics and has been widely studied over the last decade [1]. In conventional materials with high symmetry, a charge current density \( j_c \) is converted into a spin current density \( j_s = j_c \times s \), where \( s \) is the spin polarization direction) that is perpendicular to \( j_c \) and \( s \) via the spin Hall effect (SHE). In two-dimensional systems without structural inversion symmetry, the Edelstein effect (EE) emerges. In this case, an in-plane \( j_c \) leads to a perpendicularly polarized spin density \( n_s \), also in the device plane [2]. Both effects obey reciprocity, implying that the inverse transformations (of \( j_c \) and \( n_s \) into \( j_c \)) also occur with the same efficiency in the linear response regime [1]. While the SHE has been used to switch [3, 4] and probe [5, 6] the magnetization of adjacent ferromagnets, the integration of spintronic devices into logic circuits still requires the introduction of materials with better SCC efficiency. Furthermore, the search for new spin manipulation approaches requires versatile materials that allow SCC of spins polarized along different directions, a feature that is not achievable in conventional materials.

In this context, layered materials emerge as a versatile platform for efficient SCC [7–20] which induces large spin–orbit torques (SOTs) in different directions [21, 22] that can switch the magnetization of adjacent ferromagnets [23–25]. In particular, additional SCC components emerge in layered materials with reduced symmetry, such as 1T’-MoTe₂ [26, 27] and T₂-WTe₂ [21]. In van der Waals heterostructures where these materials are combined with graphene, an additional SCC component has also been observed with \( s \) parallel to the generated \( j_c \) [28, 29]. Because this component is incompatible with the bulk symmetries of MoTe₂ and WTe₂, its origin remains elusive [28].

Additionally, SOTs measured in NbSe₂/Pt bilayers have also shown the presence of an additional component not allowed by symmetry, that is sample-dependent and induced by out-of-plane...
spins parallel to $j_y$ [22]. This SCC component is not allowed due to the two mirror symmetries of bulk 2H-NbSe$_2$. These symmetries only enable the conversion of spins which are perpendicular to both $j_x$ and $j_y$ via the SHE [30–33]. Accordingly, new experiments combining graphene and van der Waals materials with high spin–orbit coupling (SOC) are required to understand the origin of the unexpected components. Furthermore, efficient conversions are expected from first-principles calculations, that have extracted spin–orbit proximities up to 40 meV in graphene/NbSe$_2$ heterostructures [34], further motivating these experiments. Finally, even though unconventional SCC components have been observed, the achievement of omnidirectional SCC in a single device has not been realized yet.

Here, we perform nonlocal spin precession experiments to investigate SCC in graphene/NbSe$_2$ van der Waals heterostructures (figure 1(a)). Our experiments show that all three spin directions ($x$, $y$, and $z$) are converted into a charge current simultaneously while keeping the $j_z$ direction fixed (see figure 1(b)) and constitute the first realization of omnidirectional SCC. Quantitative data analysis indicates that the effects originate from at least three different SCC phenomena stemming from the SOC of NbSe$_2$, its proximity with graphene and the broken symmetry at the twisted graphene/NbSe$_2$ interface.

2. Experimental details

The graphene and NbSe$_2$ flakes were prepared using the conventional mechanical exfoliation technique [35] from highly oriented pyrolytic graphite and bulk NbSe$_2$ provided by HQ Graphene. The heterostructure was prepared using the PDMS-based viscoelastic transfer technique [36] in an inert atmosphere. The Ti (5 nm)/Au (120 nm) and spin-polarized TiO$_x$ (0.3 nm)/Co (35 nm) electrodes were defined using e-beam lithography and deposited using e-beam and thermal evaporation (see supplementary information section S1). An optical microscope image of sample 1 after fabrication is shown in figure 1(a), where the used electrodes are numbered. The results shown here are obtained from sample 1, see the supplementary information section S9 for sample 2. To optimize and keep the spin transport properties of the graphene channel nearly constant with the temperature, we have tuned the carrier density far from the charge neutrality point using a backgate voltage [35]. No SCC signals have been measured near the charge neutrality point (see supplementary information sections S2, S3 and S6 for details on the $V_{bg}$ dependence of charge and spin transport in graphene). Additionally, to minimize background effects and exclude even harmonics from our measurements, we have used the DC reversal technique with an applied current of 60 µA.

3. Results

To measure SCC, we use the nonlocal measurement technique, that avoids spurious effects related to local techniques such as the Oersted fields present in SOT experiments [37] and the voltages induced by stray fields measured in potentiometric measurements [38, 39]. By applying a charge current ($I$) between electrodes 3 and 5, we inject a spin-polarized current in the graphene channel under electrode 3, leading to a pure spin current that diffuses to the NbSe$_2$-covered region, where it can get converted into a $j_z$ via SCC in the proximitized graphene and/or absorbed by the NbSe$_2$ flake in which the SCC can subsequently occur. The $j_z$ induced by SCC is detected as an open circuit voltage $V$ between the non-magnetic contacts 7 and 6, giving rise to a nonlocal signal $R_{nl} = V/I$. Due to shape anisotropy, the easy axis of our Co electrodes is along $y$. Hence, the application of a magnetic field ($B$) of more than 50 mT along $±y$ leads to the alignment of the electrode magnetization (figure 1(b)). By preparing the magnetization of electrode 3 ($M_3$) along $+y$ $≡$ $↑$ or along $-y$ $≡$ $↓$ and applying $B$ along $±x$ ($B_x$, see figures 1(c) and (d)), we obtain the nonlocal signals $R_{nl}^{x+}$ and $R_{nl}^{x-}$, respectively, which are shown in figure 2(a). To separate between the different SCC components, we define $R_{SCC} = (R_{nl}^{x+} − R_{nl}^{x−})/2$ (figure 2(b)), which contains the $y$- and $z$-SCC signals, and $R_{avg} = (R_{nl}^{x+} + R_{nl}^{x−})/2$.
Figure 2. Nonlocal spin-to-charge conversion at 100 K and \( V_{bg} = 50 \, \text{V} \). (a) \( R_{nl} \) as a function of \( B_x \) measured at the ↑ and ↓ magnetization configurations. (b) \( R_{SCC} = (R_{nl}^\uparrow - R_{nl}^\downarrow)/2 \) extracted from panel (a). (c) Antisymmetric component of \( R_{SCC} \) vs \( B_x \) corresponding to \( z \)-SCC with its associated amplitude \( \Delta R_{anti}^{SCC} \). (d) Symmetric component of \( R_{SCC} \) vs \( B_x \) corresponding to \( y \)-SCC with its associated amplitude \( \Delta R_{sym}^{SCC} \). (e) \( R_{nl} \) as a function of \( B_y \). The jumps in \( R_{nl} \) correspond to switches of the injector magnetization originated from \( y \)-SCC. (f) \( R_{avg} = (R_{nl}^\uparrow + R_{nl}^\downarrow)/2 \) extracted from panel (a) with its associated amplitude \( \Delta R_{avg}^{SCC} \).

The black line corresponds to the magnetization behavior extracted from the reference Hanle precession data. An offset of 1.85 m\( \Omega \) has been subtracted from panels (a), (e) and (f).

(figure 2(f)), which corresponds to \( x \)-SCC. Here, to simplify our notation, we refer to each SCC component as \( x \), \( y \) or \( z \)-SCC depending only on the spin direction.

First, we focus on the \( y \)- and \( z \)-SCC components. When \( B_y \) is applied, the injected spins start to precess in the \( y-z \) plane, leading to a net out-of-plane spin accumulation (\( \mu_{sz} \), see figure 1(c)). Because reversing the sign of \( B_y \) leads to opposite \( \mu_{sz} \), the \( R_{nl} \) expected from the \( z \)-SCC is antisymmetric with respect to \( B_y \). Additionally, switching (the \( y \) component of) \( M_3 \) also leads to a sign change of the signal. Accordingly, to extract the \( z \)-SCC component, we have calculated the antisymmetric component of \( R_{SCC} \left( R_{anti}^{SCC} \right) \) as a function of \( B_y \). The results from figure 2(c) confirm that \( z \)-spins are converted in our system, giving rise to a maximum \( R_{anti}^{SCC} \) of 0.13 ± 0.02 m\( \Omega \). We note that \( R_{SCC} \) also contains a clear symmetric component with \( B_x \left( R_{sym}^{SCC} \right) \). This component, which is shown in figure 2(d), might correspond to a conventional Hanle spin precession measurement, that is induced by a \( y \)-SCC component. Since the detected spins are parallel to the injected ones, the resulting \( R_{SCC} \) decreases as the spins precess towards ±\( z \) and the \( y \)-spin-projection decreases symmetrically with \( B_y \). We realize that \( B_y \) also induces the pulling of \( M_3 \) along \( x \), leading to a decrease of the signal as \( |B_y| \) increases. Because of the short spin lifetime of
our graphene channel (see supplementary information section S3), we find that the lineshape is actually dominated by contact pulling, and not by spin precession. We finally note that the apparently flat trend of figure 2(d) near \( B_x = 0 \), that is not expected from Hanle spin precession, is caused by the symmetrization of the noise in \( R_{SCC} \). We note that the \( y \)-SCC component is not expected from 2H-NbSe\(_2\) and, to confirm that this SCC component is indeed present in our sample, we apply a magnetic field along \( y \) (\( B_y \)) to control the electrode magnetization. When \( M_3 \) switches, the \( y \)-SCC signal reverses sign, allowing us to extract the signal magnitude (figure 1(b)). In figure 2(e), we show the \( B_y \)-dependence of \( R_{avg} \). When sweeping \( B_y \) from \(-75 \) to \(+75 \) mT, we observe a clear jump in \( R_{avg} \) for \( B_y \equiv B_y'' = 50 \) mT, which is caused by the switch of \( M_3 \). Additionally, sweeping \( B_y \) from \(+75 \) to \(-75 \) mT leads to a jump in \( R_{avg} \) at \( B_y \approx -B_y'' \), as expected for the switching of \( M_3 \) in the opposite direction. Note that, because \( R_{SCC} = (R_{avg}^+ - R_{avg}^-)/2 \), the spin signal in figure 2(d) should be approximately half of the 0.34 ± 0.03 m\( \Omega \) extracted from figure 2(e).

In agreement, we observe that the signal in figure 2(d) is of 0.16 ± 0.02 m\( \Omega \). Both signals are determined for \( B = 0 \).

As \( B_y \) increases, \( M_3 \) gets pulled towards \( x \), leading to the injection of \( x \)-spins (figure 1(d)). Unlike the previous spin injection components, \( x \)-SCC does not depend on the initial orientation of \( M_3 \) in the \( y \) axis. Hence, we isolate \( x \)-SCC by determining \( R_{avg} \). As shown in figure 2(f), purple dots, the signal is anti-symmetric and saturates at \( R_{avg} = 0.27 ± 0.06 \) m\( \Omega \) for \( |B_y| > 0.2 \) T, as expected from the contact magnetization behavior \([40]\). To compare \( R_{avg} \) with the \( x \)-component of \( M_3 \) (\( M_3 \sin(\theta_M) \), where \( \theta_M \) is the magnetization angle with respect to the easy axis) extracted from spin precession in the pristine graphene region (see supplementary information section S3), we have re-scaled \( \sin(\theta_M) \) and plotted it as a black line in figure 2(f). The overlap between both curves confirms that \( R_{avg} \) follows \( \sin(\theta_M) \). However, the conventional Hall effect in the graphene channel induced by the stray fields from the ferromagnetic spin injector can also lead to similar signals \([9, 41]\). To confirm that \( R_{avg} \) is induced by SCC, we have measured \( R_{SCC} \) in sample 2 as a function of an out-of-plane magnetic field to induce in-plane spin precession, an unequivocal proof for spin transport (see supplementary information section S9).

Finally, to compare between the different signals, in figure 2 we define the signal amplitudes \( \Delta R_{avg} \), \( \Delta R_{sym} \) and \( \Delta R_{anti} \), as the semi-difference between the maximum and minimum signal vs \( B_y \).

To further understand the measured SCC, we study the temperature \( T \) dependence of \( \Delta R_{avg} \), \( \Delta R_{sym} \) and \( \Delta R_{anti} \), corresponding to \( x \), \( y \) and \( z \)-SCC, respectively. In figure 3(a), we observe that \( \Delta R_{avg} \) and \( \Delta R_{sym} \) increase with \( T \) (with the exception of \( \Delta R_{avg} \) at 300 K), in contrast with \( \Delta R_{anti} \), that shows a maximum at 100 K and decreases for higher \( T \) (see supplementary information section S4 for the complete dataset and S5 for the reciprocity experiments that confirm that our work is in the linear response regime).

4. Discussion

After showing that \( x \)-, \( y \)-, and \( z \)-SCC occur simultaneously by spin precession measurements and how the resulting signals evolve with \( T \), now we proceed to discuss the origin of the different SCC components.

For this purpose, and because shunting prevents the accurate determination of the relevant transport parameters of proximitized graphene, we estimate the lower bound of the absolute value of the \( x \)-, \( y \)-, and \( z \)-SCC-associated spin Hall angles (\( \langle |\theta_{ij}^\alpha| \rangle \min \) where \( i, j, k \) and \( \alpha \) are the directions of \( j_x, j_k \) and \( s \), respectively) assuming that the SCC occurs in the NbSe\(_2\) flake (see supplementary information section S8 for details). The results from our analysis are shown in figure 3(b) as a function of \( T \). We observe that the error range (shaded areas) increases dramatically upon cooling below 100 K. This effect is caused by the decrease in \( \rho_{NbSe_2} \) (that is assumed to be isotropic) shown in the inset. The decrease in the device resistance with decreasing \( T \) leads to much smaller signals while keeping \( \langle |\theta_{ij}^\alpha| \rangle \min \) constant, decreasing the sensitivity of our measurement below 100 K. We stress that the resulting \( \langle |\theta_{ij}^\alpha| \rangle \min \) values are only valid under the assumption that the conversion occurs in the NbSe\(_2\).
flake. If the SCC occurs in the proximitized graphene, lower efficiencies are to be expected.

First, we discuss the $z$-SCC component. It can be caused by the inverse SHE in proximitized graphene [8, 10, 13, 14] (figure 4(c)), by the conventional inverse SHE in NbSe$_2$ due to a spin current polarized along $z$ and diffusing along $x$ (figure 4(f) light red and blue spins), or due to the unconventional out-of-plane SCC component in NbSe$_2$ (figure 4(f) red and blue spins) reported in [22]. The $z$-SCC origin can be discerned by changing the sign of $j_z$ in the graphene channel (along $x$-axis). If the SCC occurs in the proximitized graphene channel or by conventional SHE in NbSe$_2$, because the relevant $j_z$ flows along $x$, $R_{SCC}$ must change sign with $j_z$. In contrast, if the SCC occurs via unconventional out-of-plane SCC in the NbSe$_2$ flake, because the relevant $j_z$ points along $z$ in the NbSe$_2$, the sign of $R_{SCC}$ should not change when changing the in-plane $j_x$ direction. To discern between these effects, we compare between $R_{sd}$ obtained using electrodes 3 and 2 to inject spins (figure 4(g)). The results are shown in figures 4(h) and (i), respectively. We observe that $R_{SCC}^{xy}$ and $R_{avg}$ do not change sign when changing the spin injector (i.e. the in-plane $j_x$ direction) but $R_{SCC}^{zy}$ does, indicating that the $z$-SCC is induced by the inverse SHE in the proximitized graphene channel or the NbSe$_2$ flake. Now we need to discern between both possibilities. Looking at the spin Hall angle, we note that $\theta_{xy}^z$ is higher than 50% at 100 K. The quantification performed here assumes that $j_z$ is absorbed all along $z$ in the NbSe$_2$. Because the $x$-component of $j_z$ in NbSe$_2$ is smaller than the total $j_z$, the red line in figure 3(b) provides an even lower bound to $|\theta_{xy}^z|$ and the true spin Hall angle is expected to be higher. In this context, since 50% is already significantly larger than observed previously in NbSe$_2$ devices [22], it is safe to assume that the $z$-SCC component originates from the proximitized graphene layer instead of the NbSe$_2$. Due to shunting by the NbSe$_2$, we cannot obtain the charge and spin transport parameters of the proximitized graphene region. Since these parameters are required to quantify the proximity-induced SCC efficiency, the subsequent quantitative analysis (figure 3(c)) assuming that the SCC occurs in the NbSe$_2$ channel is not performed for the $z$-SCC component.

Next, we address the in-plane SCC components. $x$-SCC can be induced by both the conventional EE in the proximitized graphene (figure 4(a)) and the conventional SHE in NbSe$_2$ (figure 4(d)). The lack of a sign reversal of the $x$-SCC component when reversing the spin current direction (figures 4(g)–(i)) is consistent with both possibilities. As a consequence, we cannot discern between them by symmetry considerations. In bulk NbSe$_2$, the $y$-SCC component should in principle be forbidden by symmetry [22]. Nevertheless, at the first NbSe$_2$ layer, which has $C_{3v}$ symmetry,
there is an allowed SHE component that would convert y-spins propagating along x into a charge current along y [30–33]. However, the lack of a sign reversal of the y-SCC component when reversing the spin current direction (figures 4(g)–(i)) rules out this possibility. Additionally, we note that for any twist angle between the crystal mirrors of the NbSe₂ and graphene flakes that is not a multiple of 30°, the graphene and NbSe₂ vertical mirrors are not aligned, so that neither can be a mirror for the whole structure, resulting in a C₃ point group that does not have any mirror symmetry, enabling y-SCC via unconventional EE in the proximitized graphene [42, 43] as shown in figure 4(b). In contrast, when the twist angle is exactly 0 or a multiple of 30° the heterostructure belongs to the C₃v point group, which has two mirror symmetries and does not allow for the unconventional EE component (see supplementary information section S11 for a detailed symmetry discussion). Furthermore, recent first-principles calculations on twisted graphene/transition metal dichalcogenide heterostructures [44, 45] have shown that the radial component of the in-plane SOC can have a similar magnitude as the conventional Rashba SOC component [44, 46, 47] depending on the electric field and twist angle, indicating that the measured component is likely to arise from the EE in the graphene channel. Finally, shear strain could also break enough symmetries in the NbSe₂ flake, enabling a SCC component where y-polarized spins flow in the z direction in NbSe₂ (figure 4(e)) and propagate into the graphene channel [28]. The lack of a sign change of $σ_{\text{SCC}}^y$ when reversing the spin current direction (figures 4(g)–(i)) is compatible with the two mentioned mechanisms.

To gain insight into the origin of the in-plane SCC components, we discuss the $\theta^y_{\text{min}}$ shown in figure 3(b) that assumes that the SCC occurs in the NbSe₂ flake. We observe that, at 100 K, in-plane $\theta^y_{\text{min}}$ reaches the highest value, which is 25% and 20% for x- and y-SCC, respectively. At higher T, the measured signals increase, leading to minimum $\theta^y_{\text{min}}$ values of 10% and 15% at 300 K for spins polarized along x and y, respectively.

We can compare our results with SOT experiments by calculating the spin Hall conductivity $σ^y_{\text{SH}} = (\theta^y_{\text{min}} / \rho_{\text{NbSe₂}})$. We find that, at 300 K, $σ^x_{\text{SH}} = 560 \pm 200$ and $σ^y_{\text{SH}} = 850 \pm 200$ [h/e] (Ω cm)$^{-1}$. These values, which are already large at 300 K, increase an order of magnitude upon cooling to 100 K. Note that the extracted $σ^y_{\text{SH}}$ are one order of magnitude larger than the conductivities of up to 75 [h/e] (Ω cm)$^{-1}$ extracted from SOT experiments [22]. The large difference between our results and those of [22] suggests that both in-plane SCC components, that have similar magnitude, occur in the proximitized graphene layer rather than in the bulk NbSe₂. Note that, in this case, the injected spins can be converted without overcoming the interface resistance with the NbSe₂ flake (see supplementary information section S7 for the determination of the interface resistance). Accordingly, the conversion efficiency required to explain the measured signals is expected to decrease significantly. Furthermore, the reproducible observation of the unconventional y-SCC component in graphene-based heterostructures containing different layered metals [28, 29] suggests that breaking of the symmetry at the interface is the most plausible source for such a conversion.

Since the strain has been shown to fluctuate randomly in graphene-based van der Waals heterostructures [48, 49], the consistent occurrence of shear strain in all the devices seems less likely than an imperfect alignment effect. It is also worth noting that, unlike the case of graphene/semiconducting transition metal dichalcogenide devices, the samples mentioned here are not annealed at high temperatures, minimizing the probability of crystallographic alignment between the different materials [50]. Despite not having an annealing step, we have found that the interface resistances between the graphene and NbSe₂ flakes are lower than 200 Ω in the reported devices, demonstrating that the interface is transparent enough to induce proximity on the graphene flake (see supplementary information section S7 for details). For all these reasons, we believe that the signal quantification in figures 3(b) and (c) most likely does not reflect the actual SCC efficiency in our device. However, it shows that proximitized graphene allowed us to measure larger SCC signals than bulk NbSe₂ alone.

To infer whether the twist angle between the graphene and NbSe₂ flakes is a multiple of 30°, an alignment that would be incompatible with our interpretation, we have assumed that the straight edges of the flakes correspond to crystallographic directions and used the optical microscope images to estimate the twist angle between the graphene and NbSe₂ flakes in sample 1. The result is 89° ± 0.6° (see supplementary information section S10) and, even though this angle is rather small (it is only 1° from a high-symmetry point), first-principles calculations predict that the radial component of the spin texture depends more strongly on band alignments and electric fields than on the twist angle [44]. As a consequence, we believe that the small twist angle does not contradict the broken-symmetry interpretation.

Finally, we argue that the T-dependence of the SCC signal is consistent with our interpretation. Even though the x- and y-SCC signals increase with T, in contrast with the EE in graphene proximitized by a semiconductor [9], this different behavior can be attributed to the role of NbSe₂ as a shunting layer. As shown in the inset of figure 3(b), $\rho_{\text{NbSe₂}}$ increases dramatically with T, reducing the shunting and thus increasing the SCC signal. In contrast, the z-SCC component decreases when increasing T. Theoretically, it has been predicted that the SHE in proximitized graphene depends on the intervalley scattering time [51–53]. In this context, the metallic NbSe₂ flake
may induce extra intervalley scattering and have a detrimental effect on the $z$-SCC signal at high $T$.

5. Summary

In summary, we have shown that graphene/NbSe$_2$ van der Waals heterostructures convert $x$, $y$, and $z$ spins simultaneously. By analyzing the magnitude, $T$-dependence and symmetry of the SCC signals, we argue that the three components are likely to occur at the NbSe$_2$-proximitized graphene. In particular, the $z$-SCC component most likely arises due to SHE, the $x$-component due to the EE, and the $y$-SCC component due to unconventional EE at the twisted graphene/NbSe$_2$ heterostructure. A twist angle between the flakes can break all the mirror symmetries and enable the unconventional EE component. Our discovery of omnidirectional SCC paves the way for novel spintronic devices that use the three spin directions to realize complex operations. For instance, spins in different directions can be controlled independently via SOC-induced spin precession [54] and contribute to the output signal, enabling the realization of new spin-based operations.

Note added: We recently became aware of related experimental works showing omnidirectional spin-to-charge conversion in graphene/WTe$_2$ [55] and conventional SCC in graphene/NbSe$_2$ heterostructures [56].

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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ORCID iDs

Josep Ingla-Aynés https://orcid.org/0000-0001-9179-1570
Franz Herling https://orcid.org/0000-0002-4304-8173
C K Safeer https://orcid.org/0000-0003-2396-9730
Fernando de Juan https://orcid.org/0000-0001-6852-1484
Luis E Hueso https://orcid.org/0000-0002-7918-8047
Marco Gobbi https://orcid.org/0000-0002-4034-724X
Félix Casanova https://orcid.org/0000-0003-0316-2163

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