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Designing spin-textured flat bands in twisted graphene multilayers via helimagnet encapsulation

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1. Introduction

Magnetic van der Waals materials have become a fundamental building block in artificial heterostructures [1, 2]. Their two-dimensional nature provides a versatile platform to electrically control magnetism [3–6], design magnetic tunnel junctions [7–10], create artificial magnetic structures [11–13], and topological superconductivity [14, 15]. Magnetic encapsulation with van der Waals materials further provides a knob to engineer artificial quantum states [16–24]. Van der Waals helimagnets, as realized in transitional metal dihalides [25–28], provide a direction to exploit non-collinear magnetism in van der Waals heterostructures. In particular, non-collinear magnetism associated to two-dimensional multiferroics such as NiI$_2$ [29] can potentially lead to new strategies to control van der Waals magnetism electrically [30–32].

Twisted graphene multilayers have become paradigmatic systems to engineer flat bands, leading to a variety of unconventional many-body states [33–46]. The role of encapsulation in twisted graphene multilayers, in particular with boron nitride, has been shown to have a critical role in promoting specific correlated states such as Chern insulating states in twisted bilayers [43] and superconductivity in graphene trilayers [47]. Spin–orbit coupling proximity effects have also been exploited to promote specific correlated states [48, 49], and its combination with magnetic encapsulation has been shown to promote unconventional symmetry broken states [21]. However, proximity effect to non-collinear van der Waals materials has remained unexplored, especially as a means of controlling twisted van der Waals quantum states.

Here we put forward helimagnetic encapsulation as a powerful tool to control flat bands in twisted graphene heterostructures. In particular, we show that quasi-flat bands of twisted bilayer graphene (TBG) can be imprinted with strong spin textures purely by exchange coupling with helimagnets. Such spin textures do not rely on any kind of spin–obit coupling effect in graphene, providing a complementary strategy to spin-texture imprinting with spin–orbit coupling proximity. We show...
the emergence of spin-textured bands for short and long helimagnetic wavelengths and different commensuration with the graphene sublattice structure, demonstrating the robustness of helimagnet spin imprinting. We finally discuss how helimagnet imprinting impacts possible superconducting states in the flat band systems. Our results promote helimagnet encapsulation as a new strategy to design unconventional correlated states in twisted van der Waals heterostructures.

2. Model

We consider twisted bilayer graphene at a twist angle 1.44°, slightly above the flat band regime [33, 34, 50]. In this regime, the electronic structure of TBG shows strongly renormalized Dirac cones [50] and isolated moire energy bands [33, 34]. We consider TBG encapsulated between helimagnets with in-plane magnetization as shown in figure 1(a). Such heterostructure is expected to be stable as graphene shows stability for a large variety of substrates, including hBN [51, 52] WSe2 [33, 54], and the magnetic insulator Gd3 [35]. The impact of the helimagnet encapsulation is accounted by integrating out the degrees of freedom of the helimagnets, leading to an effective exchange field in the twisted graphene bilayer. The effective Hamiltonian of the proximitized multilayer takes the form

\[
H = \sum_{l} \sum_{i,j} \sum_{\alpha} \sum_{\alpha'} t_{l,i,j}^\dagger c_{l,i,\alpha}^\dagger c_{l,j,\alpha'} + \sum_{j \neq l'} \sum_{i,j} t_{l,l'}(i,j) c_{l,i,\alpha}^\dagger c_{l',j,\alpha'} + \sum_{l} \sum_{i} \sum_{\alpha,\beta} M_l(i) \cdot \sigma_{\alpha\beta} c_{l,i,\alpha}^\dagger c_{l,i,\beta} \quad (1)
\]

where \(l = 1, 2\) is the layer index, \(i, j\) are site indexes, and \(\alpha, \beta\) are spin indexes. \(t\) and \(t_{l,l'}\) are intra- and inter-layer hopping in TBG, \(\langle i, j \rangle\) restricts the sum to nearest neighbours in the first term. The interlayer hopping takes the form \(t_{l,l'}(i,j) = t_0 \left(\frac{\xi_{l,l'}(i,j) - \xi_{l,l'}(i,j)}{d_{l,l'}}\right) \exp\left[-\xi_{l,l'}(i,j)\right]\) [33, 56], where \(d\) is the distance between layers and \(\xi\) parameterizes the decay of the interlayer hopping [57]. \(J\) is the exchange coupling between the TBG and the helimagnets, \(M_l(i)\) is the magnetization around site \(i\) in the \(l\)th layer, and \(\sigma\) is a vector composed of Pauli matrices. As a reference, the hopping constants for graphene are \(t \approx 3\text{ eV}\) and \(t_{l,i}^\dagger \approx 500\text{ meV}\) [58].

The structure of the twisted multilayer is built as follows. We take \(a_{1,2}\) as lattice vectors of the bottom layer graphene: \(a_1 = \sqrt{3}a(1, 0)\) and \(a_2 = \frac{\sqrt{3}a}{2}(1, -\sqrt{3})\), where \(a\) is the lattice constant. The reciprocal vectors of graphene are then \(n_1 = \frac{1}{3a}(\sqrt{3}, 1)\) and \(n_2 = -\frac{1}{3a}(0, 1)\). The moire superlattice vectors are then given by [56]: \(A_1 = (m + 1)a_1 + ma_2\) and \(A_2 = (2m + 1)a_1 - (m + 1)a_2\), where \(m\) is an integer and in our case we take \(m = 11\). The reciprocal vectors of the moire superlattice are \(b_1 = \frac{a}{3m} + \frac{a}{2(2m + 1)}\) and \(b_2 = \frac{a}{3m} + \frac{a}{2(2m + 1)}\).

We consider a general helimagnetic order that can be incommensurate with the graphene sublattice structure. For the sake of concreteness, we consider when the magnetization on sublattice \(B\) has a relative rotation \(\theta_0\) w.r.t. that on sublattice \(A\)

\[
M(i \in A) = M_0(\cos(\mathbf{q} \cdot \mathbf{R}_l), \sin(\mathbf{q} \cdot \mathbf{R}_l), 0)
\]

\[
M(i \in B) = M_0(\cos(\mathbf{q} \cdot \mathbf{R}_l + \theta_0), \sin(\mathbf{q} \cdot \mathbf{R}_l + \theta_0), 0)
\]

with \(M_0\) being the magnitude of the local magnetization, \(\mathbf{q}\) being the characteristic wave vector of the helimagnet, and \(\mathbf{R}_l\) being the coordinate of site \(i\). Due to the superexchange mechanism [59], the local magnetization at top and bottom magnets are expected to align antiferromagnetically, so we consider \(M_1(i) = -M_2(i) = M(i)\). For the sake of simplicity, we consider \(\mathbf{q}\) parallel to \(\mathbf{b}_1 - \mathbf{b}_2\), which is a high-symmetry direction of the moire Brillouin zone. When \(\theta_0 = 0\), there is no sublattice imbalance and the magnets are fully characterized by the helimagnetic order (figure 1(b)). When \(\theta_0 = \pi\), the helimagnetic order is overlaid with a staggered magnetization for the sublattices.

At low energy, the influence of the helimagnetization on the band structure of TBG depends only on the effective exchange field \(J_0 = JM_0\) and the ratio between the characteristic vector \(\mathbf{q}\) and the moire periodicity. Given that the moire periodicity can be tuned by the twist angle \(\theta\), which can be controlled by tear and stack fabrication [60, 61], we explore the following two regimes: (i) the helimagnet periodicity is an integer times the periodicity of the moire lattice, denoted as a commensurate regime, or (ii) the helimagnet periodicity is much smaller than the periodicity of the moire lattice, which we denote as an incommensurate regime. We show that in both cases the helimagnetization imprints a spin texture in quasi-flat bands in TBG.

In the commensurate regime, the system can be solved with equation (1). In the incommensurate regime, however, the whole system no longer has the periodicity of the moire lattice, and we adopt the generalized spinor-Bloch theorem to diagonalize the system [62–64]. We perform a local unitary transformation to the Hamiltonian such that the local magnetization is aligned along the \(x\) direction for all sites [63, 64]:

\[
U = \prod_i e^{-i\xi_{\sigma,s}^x} \quad (3)
\]
with $\sigma_{x,i}$ the spin Pauli matrix in site $i$. The transformed Hamiltonian becomes:

$$H' = U^\dagger H U$$

$$= \sum_{l} \sum_{(i,j)} \sum_{\alpha,\beta} t_{l} e^{i\theta(i,j)}(\sigma_{x})_{\alpha\beta} c_{l,\alpha,i}^\dagger c_{l,\beta,j}$$

$$+ \sum_{l \neq l'} \sum_{(i,j)} \sum_{\alpha,\beta} t_{l} e^{i\theta(i,j)}(\sigma_{x})_{\alpha\beta} c_{l,\alpha,i}^\dagger c_{l',\beta,j}$$

$$+ J_0 \sum_{l} \sum_{i \in \{A\}} \sum_{\alpha,\beta} f_{l} c_{l,\alpha,i}^\dagger (\sigma_{x})_{\alpha\beta} c_{l,\beta,i}$$

$$+ J_0 \sum_{l} \sum_{i \in \{B\}} \sum_{\alpha,\beta} f_{l} c_{l,\alpha,i}^\dagger (\sigma_{x})_{\alpha\beta} c_{l,\beta,i}$$  \hspace{1cm} (4)

where $J_0 = |M_0|$ is the effective local exchange field, $f_1 = -f_2 = 1$, $\theta(i,j) = \mp \frac{1}{2} q \cdot (R_i - R_j)$ and $\chi = \cos \theta_0 = \pm 1$ for $\theta_0 = 0, \pi$. Since the magnetization is uniform up to a sublattice imbalance in $H'$, $H'$ has the same periodicity as an isolated TBG. With no proximity effect, i.e. when $J_0 = 0$, the only difference between $H$ and $H'$ is the additional phases in the hopping terms, resulting in a momentum shift of $\pm q/2$ for spin-up/down channels, respectively (figure 1(c)). We note that the additional phase in the first two terms correspond to an artificial inplane spin–orbit coupling, while the last two terms correspond to exchange terms. For finite $J_0$, both time-reversal symmetry and inversion symmetry are broken in $H$, and spin-mixing occurs in the quasi-flat bands, creating a spin texture (figure 1(d)). We note that the broken symmetries stem solely from the helimagnetic encapsulation, and do not require external magnetic field or substrate. In the following we address the spin-textured quasi-flat bands in TBG due to the proximity to the helimangets for both commensurate and incommensurate helimagnets.

3. Electronic structure with commensurate helimagnetic encapsulation

We start with the simplest case when the helimagnets are commensurate with the TBG supercell and the band structure can be computed with the Hamiltonian in equation (1). When $J_0 > t_{1}^{d}$, the exchange coupling will be much larger than the bandwidth of the quasi-flat bands in TBG, and will substantially change the bands. Therefore we consider $J_0 < t_{1}^{d}$, which corresponds to a realistic regime with proximity to insulating magnets \cite{65}. We note that in this regime, the Hamiltonian can be solved without including a spin rotation in the Bloch phase, as the Hamiltonian in the original basis has the periodicity of the moire supercell. We first comment on the case $\theta_0 = \pi$. The spin spiral field acts as an artificial

Figure 1. (a) Sketch of TBG encapsulated with helimagnets, viewed from top and side. (b) Local effective exchange field induced by proximity to helimagnets on top (red) and bottom (blue) layers of TBG. The direction of the characteristic vector of the helimagnets $\mathbf{q}$ is denoted with the black arrow. (c) Band structure of the heterostructure described by equation (4) with $J_0 = 0$ and $q = 0.05(\mathbf{b}_1 - \mathbf{b}_2)$. (d) Band structure of the heterostructure with $J_0 = 0.033t_{1}^{d}$, $\theta_0 = \pi$ and $q = 0.05(\mathbf{b}_1 - \mathbf{b}_2)$. We took twist angle 1.44° for (c), (d).
spin–orbit coupling, generating a spin-splitting in momentum space. Interestingly, in this regime, the band structure does not exhibit sizable anticrossings driven by $J_0$. This behavior stems from the orthogonality between low energy states, due to the $\pi$ Berry phase of the Dirac cones \cite{58}. Since the low energy states are orthogonal, a coupling between these states does not result in a splitting. Therefore, the splitting, to the lowest order of $J_0$, comes from the coupling between the low energy bands with higher energy bands, and has a quadratic dependence on $J_0$. In the small $J_0$ regime we considered, a sizable gap will not open.

We now focus on the case of $\theta_0 = 0$, which leads to a strong change in the electronic structure induced by the helimagnetic field. The band structure along $b_1 - b_2$ for different values of $q$ and $J_0$ are shown in figures 2(a)–(d). We see that spin-splitting occurs in momentum space, stemming from the spiral field. In addition, the band structures exhibit anticrossings between the spin-channels, stemming from a first-order contribution in $J_0$ to the electronic energies. We note that both the spin-splitting and the anticrossings become smaller for larger $q$.

To investigate the impact of the helimagnets on the flatness of the quasi-flat bands, we present the density of states (DOS) $\rho(\omega)$ vs $J_0$ for $q = b_1 - b_2$ (figure 2(e)). We see that as $J_0$ increases, the peaks in the DOS stemming from the quasi-flat bands split and the flat bands retain a small bandwidth. The previous phenomenology takes place...
for different values of \( q \) at \( J_0 = 0.2t_0^b \) as shown in figure 2(f). We find that the DOS increases with \( q \) and for \( q = 2(b_1 - b_2) \) the DOS is almost the same as the DOS of pristine TBG. We have thus found that the helimagnets induce spin-splitting and anticrossing in quasi-flat bands in TBG. Both effects become more significant for larger effective exchange coupling \( J_0 \) and smaller characteristic vector \( q \) of the helimagnets. In the next section, we focus on the regime when \( q \ll b_1 - b_2 \), where similar effects are expected to occur at smaller values of \( J_0 \).

4. Electronic structure with incommensurate helimagnetic encapsulation

We now move on to investigate the modification of the band structure with helimagnetization in the small \( q \) regime. In particular, we focus on \( q \leq 0.2(b_1 - b_2) \) such that the helimagnetization does not result in intervalley scattering. We consider both \( \theta_0 = 0 \) and \( \theta_0 = \pi \) cases.

For \( \theta_0 = 0 \), we have a helimagnet that induces a nearly ferromagnetic exchange in neighboring sites. In this case, the band structure with \( q = 0.05(b_1 - b_2) \) and different values of \( J_0 \) is shown in figures 3(a) and (b). Similar to the large \( q \) case discussed in section 3, spin-splittings and anticrossings appear in the nearly flat bands. The anticrossing gap \( \Delta \) at the \( K \) point as denoted in figure 3(a) exhibits a quadratic dependence on \( J_0 \), indicating that the anticrossing is caused by a second-order contribution. The reason that the first-order contribution does not cause spin-splitting is due to the orthogonality between the low energy eigenstates [38], similar to the \( \theta = \pi \) case in the commensurate \( q \) regime. Comparing the band structure with different \( q \) at \( J_0 = 0.13t_0^b \) (figures 3(b)–(d)), we find that the spin texture exhibits a strong dependence on \( q \), whereas the dispersion does not change substantially with \( q \). The DOS versus \( J_0 \) and \( q \) (figures 3(e) and (f)), show that helimagnetic encapsulation does not spoil the small bandwidth of the low energy bands. We find that as \( J_0 \) increases, the peaks in the DOS split and eventually the DOS slightly decreases. The DOS shows small \( q \)-dependence for \( q > 0.05(b_1 - b_2) \), and only when \( q \) is close to 0 the DOS decreases. In this regime, a relatively small \( J_0 \) leads to a strong spin texture in the quasi-flat bands, creating a strongly \( q \)-dependent spin texture.

We now move on to the case when \( \theta_0 = \pi \), i.e. the magnetization on the sublattices are opposite and we have a helimagnet inducing a nearly anti-ferromagnetic field in neighboring sites. The band structures with \( q = 0.05(b_1 - b_2) \) and different \( J_0 \) are shown in figures 4(a) and (b). In this regime, large anticrossings at \( K \) and \( K' \) appear, together with a simultaneous splitting of the Dirac cones. The splitting \( \Delta' \) as denoted in figure 4(a) exhibits a linear dependence on \( J_0 \), indicating that the splitting stems from a first-order contribution. As a consequence of the splitting of the Dirac cones, the bands become flatter and shift away from each other as \( J_0 \) increases, which is in contrast to the case when \( \theta_0 = 0 \). The \( q \)-dependence of the bands with fixed \( J_0 = 0.13t_0^b \) is shown in figures 4(b)–(d). Interestingly, both the spin texture and the dispersion depend strongly on \( q \). The low energy DOS versus \( J_0 \) and \( q \) is shown in figures 4(e) and (f), highlighting that the nearly flat bands maintain their flatness as the system develops a strong spin texture.

Compared to the large \( q \) regime as discussed in section 3, the small \( q \) regime with \( \theta_0 = 0 \) shows bands whose dispersion is less dependent on \( q \) whereas the spin texture is more sensitive on \( q \), making it more favorable for designing spin-textured flat bands. In addition, in the case when \( \theta_0 = \pi \), the quasi-flat bands in TBG can be further flattened with helimagnetic encapsulation. In all the regimes discussed, quasi-flat bands with spin texture can be engineered in TBG by proximity to helimagnets. We note that the results do not rely on fine-tuning the twist angle in TBG as we have considered a non-magic angle.

5. Spin-textured Fermi surfaces and superconducting instabilities

In this section we discuss the impact of the imprinted spin texture on the superconducting instability in TBG. The superconductivity in TBG can be formally derived from a full tight-binding model including long-range interactions:

\[ \mathcal{H} = H + \sum_{i,j,k,l,\alpha\beta} V_{ijkl} c_{i\alpha}^{\dagger} c_{j\beta} c_{k\alpha} c_{l\beta} \]  

where \( H \) is the Hamiltonian from equation (1), and the interaction term contains an attractive channel stemming from electron-phonon coupling or other attractive mechanisms mediated by quasiparticle fluctuations. Solving \( \mathcal{H} \) using a mean-field approximation would give rise to a superconducting term in a Bogoliubov–de-Gennes Hamiltonian. However, since in TBG the size of the minimal unit cell is on the order of thousands of sites, solving such Hamiltonian with interactions represents a remarkable problem even at the mean-field level. To overcome these limitations, here we focus our discussion on a phenomenological symmetry analysis of the superconducting state, in particular focusing on how its symmetry properties are controlled by the underlying electronic structure. Superconductivity in the weak coupling limit is understood in terms of a Fermi surface instability by the formation of Cooper pairs.
with total momentum equal to zero [66]. This means that the electrons forming the pairs have opposite momenta, and therefore belong to states that are related by inversion or by time-reversal symmetry (TRS). The breaking of these two key symmetries by the helimagnet encapsulation in principle hinders the formation of Cooper pairs. In the following we show explicitly the spin-textured Fermi surfaces of the helimagnetically-encapsulated TBG and discuss their impact on the robustness of different types of superconducting states. We consider a hole doping with two holes per moire supercell.

In the absence of exchange coupling to the helimagnets, the Fermi surfaces have six-fold symmetry around the center of the Brillouin zone and are spin-degenerate, such that all types of spin-configuration for the Cooper pairs are possible. As a reference, in the absence of the effective exchange magnetic field, the Fermi surface of the Hamiltonian in the rotated basis equation (4) with $q = 0.05(b_1 - b_2)$ and $J_0 = 0.067t_0$. Inset: spin-splitting $\Delta$ at K point. We took in (a) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.067t_0$. In (b) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (c) $q = 0$ and $J_0 = 0.13t_0$, in (d) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (e) $q = 0.05(b_1 - b_2)$ and in (f) $J_0 = 0.13t_0$.

Figure 3. (a)–(d) Band structure and (e)–(f) DOS $\rho(\omega)$ of TBG with incommensurate helimagnetic encapsulation described by equation (4). The helimagnetic order, given by equation (2), has a characteristic vector $q$ much smaller than the moire reciprocal vectors of TBG and $\theta_0 = 0$. We took in (a) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.067t_0$. Inset: spin-splitting $\Delta$ at K point. We took in (b) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (c) $q = 0$ and $J_0 = 0.13t_0$, in (d) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (e) $q = 0.05(b_1 - b_2)$ and in (f) $J_0 = 0.13t_0$. 

\[ q_{\perp} \]
Figure 4. (a)–(d) Band structure and (e)–(f) DOS $\rho(\omega)$ of TBG with incommensurate helimagnetic encapsulation described by equation (4). The helimagnetic order, given by equation (2), has a characteristic vector $q$ much smaller than the moire reciprocal vectors of TBG and $\theta_0 = \pi$. We took in (a) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.067t_0$, in (b) $q = 0.05(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (c) $q = 0$ and $J_0 = 0.13t_0$, in (d) $q = 0.1(b_1 - b_2)$ and $J_0 = 0.13t_0$, in (e) $q = 0.05(b_1 - b_2)$ and in (f) $J_0 = 0.13t_0$.

a horizontal mirror symmetry [67]. Under these circumstances, spin-singlet ($\sim |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$) and spin-triplet pairing with the d-vector along the z-axis ($\sim |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$) are stable [68–72]. The spin-mixing is also present in the Fermi surface when a commensurate helimagnet encapsulation is considered (figure 5(d)), and the above argument applies. Interestingly, as the proximity to the helimagnet introduces inversion symmetry breaking, the mixing of superconducting states of different parity is allowed [73, 74]. If the dominant pairing channel is s-wave, inversion symmetry breaking would induce a p-wave component to the superconducting gap structure due to the presence of an effective Rashba spin–orbit coupling. The converse situation is also true, if the dominant pairing channel is p-wave, some s-wave component is induced by inversion symmetry breaking. Ultimately, this allows to change the parity of the dominant channel in the superconducting gap by encapsulation with strong helimagnets. Interestingly, helimagnetic encapsulation can potentially result in Yu–Shiba–Rusinov bound states, as these usually appear around magnetic impurities. These states can be probed with tunneling spectroscopy, and provide a characterization of the underlying superconducting order [75].
Figure 5. Fermi surfaces of TBG with helimagnetic encapsulation with a doping of two holes per moire supercell, with different characteristic vector $\mathbf{q}$ and effective coupling $J_0$. We took in (a) $\mathbf{q} = 0.05(\mathbf{b}_1 - \mathbf{b}_2)$ and $J_0 = 0$, in (b) $\mathbf{q} = 0.05(\mathbf{b}_1 - \mathbf{b}_2)$, $J_0 = 0.067t^a_0$ and $\theta_0 = 0$, in (c) $\mathbf{q} = 0.05(\mathbf{b}_1 - \mathbf{b}_2)$, $J_0 = 0.067t^a_0$ and $\theta_0 = \pi$, and in (d) $\mathbf{q} = \mathbf{b}_1 - \mathbf{b}_2$, $J_0 = 0.2t^a_0$ and $\theta_0 = 0$.

$\mathbf{b} = |\mathbf{b}_1| = |\mathbf{b}_2|$ is the length of the reciprocal vectors.

6. Conclusion

To summarize, we have shown how nearly-flat bands of twisted graphene multilayers can be imprinted with non-trivial spin textures via helimagnet encapsulation. In particular, we have shown the emergence of strong spin textures both in the limit of large and small $\mathbf{q}$-vector in comparison with the moire unit cell. Interestingly, we demonstrated that the magnetic encapsulation alone is capable of opening up a gap at the Dirac points of the flat bands, maintaining the flatness of the low energy bands. We have shown that the existence of such spin textures dramatically impacts the spin structure of the Fermi surface. Finally, we discussed the potential of helimagnetic encapsulation to promote unconventional superconducting states in the system. Our results put forward helimagnet encapsulation as a powerful technique to design spin-textured flat bands and promote unconventional superconducting states in twisted graphene multilayers.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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