Linear magnetoresistance in mosaic-like bilayer graphene

Key parameters of samples presented in the main manuscript.

Table SI 1 The length denotes the distance between the inner probes in a Hall bar. The mean free path \( l \) was calculated using \( l = \frac{\hbar \mu}{e \cdot p \cdot \pi} \).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Charge carrier density ( p ) (10^{12}/cm^2)</th>
<th>Hall-mobility ( \mu ) (cm^2/Vs)</th>
<th>Mean free path ( l ) (nm)</th>
<th>Width (µm)</th>
<th>Length (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 1 a</td>
<td>6.7</td>
<td>2700</td>
<td>82</td>
<td>50</td>
<td>285</td>
</tr>
<tr>
<td>Fig. SI 1 a</td>
<td>12</td>
<td>940</td>
<td>38</td>
<td>20</td>
<td>120</td>
</tr>
<tr>
<td>Fig. 1 c</td>
<td>7.4</td>
<td>1470</td>
<td>47</td>
<td>2</td>
<td>8</td>
</tr>
<tr>
<td>Fig. SI 1 b</td>
<td>7.2</td>
<td>1460</td>
<td>46</td>
<td>74</td>
<td>1600</td>
</tr>
<tr>
<td>Fig. 1 b</td>
<td>8.7</td>
<td>1290</td>
<td>44</td>
<td>90</td>
<td>1600</td>
</tr>
</tbody>
</table>

Pulsed magnetic field data. Fig. [S1a] shows that there is no significant difference appearing between the magnitude of the resistivity resulting from 2-point and 4-point resistance measurements. The most striking feature is that the 2-point MR is symmetric with magnetic field, whereas 4-point MR shows an asymmetry due to inhomogeneities. This asymmetry also varies from sample to sample (see Fig. [S1b]). The onset of Shubnikov-de-Haas oscillations differs significantly for both samples.

Figure SI 1 Pulsed field data of samples not presented in the main manuscript. (a) and (b) Data of further samples measured in pulsed magnetic fields. The qualitative behavior of the MR is equal to that described in the main manuscript. Two-point data in (a) was calculated using an overall sample length of 155 µm.
Using a Landau-level-spectrum of bilayer graphene
\[ E_n = \hbar \omega_c \sqrt{N(N-1)} \] (1)
together with a dispersion relation
\[ E(k) = -mv_F^2 + \sqrt{(mv_F^2)^2 + (\hbar v_F k)^2} \] (2)
using \( m \approx 0.035 \, m_e \) and \( v_F \approx 10^6 \, \text{m/s} \) (see 1), one can obtain the magnetic field at which the lowest non-zero Landau-level (N=2) crosses the Fermi-energy:

<table>
<thead>
<tr>
<th>( p , (10^{12}/\text{cm}^2) )</th>
<th>( B , (T) )</th>
<th>Figure</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.2</td>
<td>68</td>
<td>Figure SI 1 (b)</td>
</tr>
<tr>
<td>8.7</td>
<td>85</td>
<td>Figure 1 (b)</td>
</tr>
<tr>
<td>12</td>
<td>121</td>
<td>Figure SI 1 (a)</td>
</tr>
</tbody>
</table>

These fields largely exceed the onset of linear MR. Hence, the quantum MR proposed by Abrikosov can be excluded in this regime\(^2\). We are convinced that these arguments also hold for the multi-layer graphene samples investigated by Friedman et al., although their charge density may be significantly smaller. In this paper, linear MR was observed even at 1 T and room temperature and was assigned to the Abrikosov mechanism. This was justified by arguing against the classical mechanism, as the scaling of the slope and the set-in of linear MR was insensitive to temperature. Note that the charge carrier mobilities in multi-layer graphene on the carbon face of SiC are known to be very high, which would justify the set-in of linear behavior at small fields. To our opinion, exactly these arguments point towards a classical mechanism. The behavior of both quantities in our samples, where quantum MR is absent, is discussed below.

**Data of multiple samples**

A linear behavior in the magnetoresistance is characterized by its slope (in addition to the axis intercept). In the theoretical description given by PL for the homogeneous network, the slope \( 1/R_0 dR(B)/dB \) is presented, which is proportional to the charge carrier mobility \( \mu \) when no contact resistances in between segments are assumed. We find it more appropriate to analyze the slope \( dR(B)/dB \), which is a function of \( 1/en \) for negative charge carriers, or \( 1/ep \) for positive charge carriers, as the Hall resistance in each segment is determined by this quantity. This quantity is insensitive to contact resistances. We present the correlation of the slope on the charge density for many samples, which gives, however, no conclusive experimental picture. This can be understood when comparing with extensive simulations of finite, disordered matrices, which show similarly strong scatter of the resulting slope.
Using a Landau-level spectrum of bilayer graphene

\[ E_n = \hbar \omega_c n \left( N - 1 \right) \]  

(1)

together with a dispersion relation

\[ E(\mathbf{k}) = -m v_F^2 k^2 + \frac{\hbar v_F^2}{2} k^2 + \frac{\hbar v_F}{2} k^2 \]  

(2)

using \( m \approx 0.035 m_e \) and \( v_F \approx 10^6 \text{m/s} \) (see 1), one can obtain the magnetic field at which the lowest non-zero Landau-level (N=2) crosses the Fermi-energy:

\[ B(10^{12} / \text{cm}^2) \]  

Figure 7.2

Figure SI 1(b)

Figure 8.7

Figure SI 1(a)

These fields largely exceed the onset of linear MR. Hence, the quantum MR proposed by Abrikosov can be excluded in this regime2. We are convinced that these arguments also hold for the multi-layer graphene samples investigated by Friedman et al., although their charge density may be significantly smaller. In this paper, linear MR was observed even at 1 T and room temperature and was assigned to the Abrikosov mechanism. This was justified by arguing against the classical mechanism, as the scaling of the slope and the set-in of linear MR was insensitive to temperature. Note that the charge carrier mobilities in multi-layer graphene on the carbon face of SiC are known to be very high, which would justify the set-in of linear behavior at small fields. To our opinion, exactly these arguments point towards a classical mechanism. The behavior of both quantities in our samples, where quantum MR is absent, is discussed below.

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Figure SI 2 Overview of measured samples. Slope of four-point magnetoresistance \( (dR(B)/dB) \) for multiple samples, plotted as a function of the charge carrier density \( p \) (reciprocal scale) obtained from a Hall-measurement. Each and every sample displayed linear MR. The data were taken in the range \(|B| \leq 8 \text{T} \) (superconducting magnet). Equal symbols and colors correspond to different samples on the same chip (i.e. equally processed). The red line shows the dependence of the slope on the charge carrier density for a PL network, for which \( 1/ep \) fully determines the slope. This result does only weakly depend on the aspect ratio of the network or the type (three-fold interconnected/four-fold interconnected networks). The difference between the network model and the measurements varies strongly for different samples. This indicates that the individual network geometry in combination with perturbations of the network (i.e. defects) determines the exact strength of the linear contribution. No further correlation between slope and sample parameters could be identified.

Figure SI 3 Magnetoresistance of a further small sample, containing only bilayer.
Extensions to the PL model

We have established a simulation following the Parish and Littlewood model, for which each segment was parametrized by a charge carrier mobility \( \mu_{n,m} \) and a charge density \( p_{n,m} \), both for a four-terminal interconnected matrix as well as a three-terminal interconnected matrix (cf. Fig. 2e in main manuscript). We analyze both the two-terminal resistance \( R_{2p}(B) \) that is linear in \( B \) in perfect agreement with the PL model as well as a four-terminal resistance \( R_{4p}(B) \) that employs two additional voltage probes at the lower (or, equivalently upper) edge. \( R_{4p}(B) \) turns out to be only very weakly linear in \( B \), close to constant, for homogeneous networks.

As our samples have predominantly structural disorder, induced by the irregular network of partial dislocations, we considered defects of the “shortcut” type, where individual discs were set to zero resistance (technically, \( p = \infty \) was chosen). These shortcuts distort the current distribution and the potential landscape. Fig. [S4a] shows the result of this simulation in a three-terminal connected network. The more defects are added, the more the slope of \( R_{4p}(B) \) and \( R_{2p}(B) \) converge. This indicates that disorder blurs the qualitative difference of 2-point and 4-point MR, which matches to the experimental findings. This underscores why the linear MR is a very robust finding in mosaic conductors, which appears independent of the contact topology.

Onset of linear regime at small fields

The linear magnetoresistance stems from a local admixture of transversal Hall currents (proportional in \( B \)) and longitudinal currents. Consequently, the transition to linear behavior occurs qualitatively, when the Hall currents exceed the longitudinal currents in a segment, that is when \( \mu_{\text{segment}}B \) equals 1\(^3\). \( \mu_{\text{segment}} \) may not be confused with the overall charge carrier mobility \( \mu_{\text{overall}} \). This has led to a confusion in the past, where the early set-in of linear behavior at values smaller than \( \mu_{\text{overall}} \) was assigned to strong mobility fluctuations. Mobility fluctuations, however, are not essential, they are just one possible source of disorder.

\( \mu_{\text{overall}} \) can be extracted from macroscopic Hall measurements (see table SI1), or from the crossing point of the long-range electron-electron interaction correction\(^5\), which gives consistent results.

However, we have traced the mean free path over ~40 samples and find consistently that the low-temperature mean free path in bilayer graphene is of the order of 50 nm. Together with the TEM observation of a network of partial dislocations, which provides segmentation on a 50 nm scale, we find that in bilayer graphene, the typical distance of dislocations is the smallest scattering length. The transport inside the segment may therefore be considered as pseudo-ballistic and dominated by boundary effects. The concept of an individual segment mobility is therefore not well defined. However, in the enforcing the link to the PL model it is evident that the properties of the individual conducting discs of that model should be those of the homogeneous bilayer, i.e. free from partials. The mobility of partial free bilayer graphene is expected to be of the order of 10,000-20,000 cm\(^2\)/Vs and this corresponds well with the cross over to the linear regime at \( B \sim 1 \) T (all numbers in this section are qualitative and may vary within a factor of less than two).
Graphene is expected to be of the order of 10,000-20,000 cm²/Vs and this corresponds well of the homogeneous bilayer, i.e. free from partials. The mobility of partial free bilayer is evident that the properties of the individual conducting discs of that model should be those of the smallest scattering length. The transport inside the segment may therefore be considered as occurring qualitatively, when the Hall currents exceed the longitudinal currents in a segment, this sets the slope of the magnetoresistance (MR) that employs two additional voltage probes at the lower (or, equivalently upper) edge. It turns out to be only very weakly linear in the range of magnetization where the MR is measured. At the same time, the low-temperature mean free path in bilayer graphene is of the order of 50 nm. Together with the TEM observation of a network of partial dislocations, which provides segmentation and may vary within a factor of less than two).

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Quantum calculations of the network wavefunction. In this section of the SI we present details of the quantum calculation of the network wavefunction. Evidently, to solve such a problem for the experimental network on a 1000 x 1000 nanometer² area is not possible with within the framework of any atomistic scheme; we therefore turn to an effective Dirac-Weyl approach. The diagonal blocks, see Eq. 3, of the required effective Hamiltonian will possess the Dirac-Weyl operators from each layer, note that the second layer Dirac-Weyl operator is conjugated. The crucial off diagonal blocks must represent a position dependent effective field that on the AB and AC regions reproduces the well known result for the low energy description of these two stackings.
\[
H_{\text{general}} = \begin{pmatrix}
0 & p_x - i p_y & S_{11}(r) & S_{12}(r) \\
p_x + i p_y & 0 & S_{21}(r) & S_{22}(r) \\
S_{11}(r)^\dagger & S_{21}(r)^\dagger & 0 & p_x + i p_y \\
S_{12}(r)^\dagger & S_{22}(r)^\dagger & p_x - i p_y & 0
\end{pmatrix}
\] (3)

Thus the effective field on the AB, AC, and AA regions (at the 6-fold intersection of partials this structure is realized, see Ref. \textsuperscript{5}), will take on (up to a phase) the following form:

\[
S_{AB} = \begin{pmatrix}
1 & 0 \\
0 & 0
\end{pmatrix} \quad S_{AC} = \begin{pmatrix}
0 & 0 \\
0 & 1
\end{pmatrix} \quad S_{AA} = \begin{pmatrix}
0 & 1 \\
1 & 0
\end{pmatrix}
\] (4)

The question is then how to interpolate between these regions, that is, the structure that this quantum field should assume on the partials of the network. This problem is reminiscent of that of solving the graphene twist bilayer in the effective field scheme. In that problem the stacking vector (i.e., the relative shift of the lattices) takes on, in the small angle limit, all possible shift vectors. Here however, the deviation from a relative translation of the layers resulting in AB or AC stacking occurs only at the partials. Following the earlier work on the twist bilayer\textsuperscript{7,8} we can derive a general formula that relates the relative displacement vector of the two layers at \( r \) to the effective field at this point.

\[
S(r) = \frac{\Omega}{(2\pi)^2} \sum_{i=1}^{3} t_K M_i e^{-i K_i \cdot \Delta u(r)}
\] (5)

This field is precisely that which should enter Eq. [3] and relates in a transparent way the structural displacement field \( \mathbf{u}(r) \) to the quantum effective interlayer field. Details of the derivation of the formula will be presented elsewhere. In Eq. 5, \( \Omega \) is the volume of the single layer graphene Brillouin zone, \( t_K \) the Fourier transform of the interlayer hopping at the K point (i.e., the vertices of the single layer graphene Brillouin zone), \( K_i \) the vectors of the three symmetry equivalent K points, and \( \mathbf{u}(r) \) the displacement field. The softness of the

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**Figure S1 5 Interlayer coupling field of the partial dislocation network.** Shown is the projection of the interlayer field given by Eq. 5 into its (a) AB (b) AA (c) AC components. The breakdown of the interlayer field in this way clearly reveals the structure of the dislocation network; note that the AB and AC fields are essentially the complement of each other, as would be expected. The AA component of the interlayer field is naturally small, and is significant only at the partials and more prominently at the 6-fold nodes of the partial network. Note also that shown here is the absolute value of the complex valued interlayer field, and thus the phase structure is not visible in these plots.
partials (a width of 5 nm is reported from experiment) allows us to exclude intervalley scattering. The matrices $M$ are given by

$$
M_1 = \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad M_2 = \begin{pmatrix} 1 & e^{i2\pi/3} \\ e^{i2\pi/3} & e^{-i2\pi/3} \end{pmatrix} \quad M_3 = \begin{pmatrix} 1 & e^{-i2\pi/3} \\ e^{-i2\pi/3} & e^{i2\pi/3} \end{pmatrix}
$$

and are identical to the matrices found in the twist bilayer formalism. The particular form assumed by these matrices depends on the initial stacking chosen before the displacement field is applied that generates the experimental partial dislocation network. We assume an initial AB stacking (this choice is of course arbitrary and any initial stacking will lead to the same $S(r)$ simply via different $M$ matrices). The structure of the displacement field we take from the experimental TEM images. For the structure of the partials we use a displacement field modeled on that found in semi-empirical atomistic calculations of a simpler single partial unit cell presented in Ref. 9. Namely, the displacement field across a partial is given by the following formula

$$
d(l) = d_i + (d_j - d_i) \eta
$$

where $l$ is measured along the normal to the dislocation and $\eta$ is given as

$$
\eta = \frac{1}{2} \left( \frac{\tanh(\epsilon l)}{\tanh(\epsilon L_2)} + 1 \right)
$$

This is a function that runs from 0 to +1 and so in conjunction with Eq. [7] results in a stacking vector that evolves smoothly from that of one grain to the other. We choose $\epsilon = 0.10$ Å$^{-1}$ and $L_2 = 50$ Å and in this way find a displacement field across the partial that closely resembles that found in the semi-empirical calculations of Ref. 9.

The interlayer field determined in this way is presented in Fig. [S5]. In this figure we present the components of the field resulting from a projection onto the matrices which describe AB, AC, and AA stacking respectively; this then yield the “relative contribution” from each of these stacking types. As one would expect the AB/AC structure of the partial network is clearly delineated as can be seen by a comparison of panel (a) of Fig. [S5], the AA component of the field, and panel (c) of Fig. [S5] the AC component. It may also be observed that the AA contribution of the field is important only on the partials and, more significantly, at the 6-fold nodes of the network.
Figure SI 6 Zoom of the node structure of the partial network. (a) AA projection of the interlayer field; note that 6-fold nodes possess an AA spot at the vertex of the node, while nodes that are 3-fold do not (there structure at the centre is an intermediate shift vector between the AB and AC structure). (b) AB projection of the same region of the partial network.

This may be seen in Fig. [S6] in which the network field is shown for a limited region of the unit cell. To construct the field at these nodes a similar formula to Eqs. [7,8] is deployed but with the distance perpendicular to the partial replaced by the radial distance from the node centre. At the 3-fold nodes the required structure is likely not to be the AA stacked structure found in the STM results for 6-fold nodes presented in Ref.5, and in the absence of experimental input we must therefore make an assumption concerning this. We choose to take the stacking vector at the centre of such a node to be a displacement vector precisely half of that to reach the AC structure from the AB; this would appear reasonable given than if one draws a line directly through the node one traverses from AB to AB stacking, in contrast to the 6-fold nodes where such a line would result in a change of stacking through the node from AB to AC.

Finally having determined the effective field that must enter Eq. [3] the effective Hamiltonian is now to be solved. Details of this procedure are presented in the methods section. However, we note here a crucial point is that to recover a periodic problem we fold the unit cell of Fig. [S5] 4-times via mirroring. This results in a periodic field, at the cost of a unit cell area increased by a factor of 4.

Network wavefunctions in a hexagonal dislocation network. In this section we present calculations for a model partial dislocation network, that of an approximately hexagonal arrangement of partials. In this network the Burgers vector at each of the three distinct directions of partials is one of the three allowed such vectors, see Figure [2] of the manuscript. The effective interlayer field is calculated in precisely the manner described above and the resulting quantum problem solved as described in Methods. In Fig. [S7] we present a series of wavefunctions at 4 energies that illustrate essentially the same physics as seen in the far more complex experimental network.
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**Figure SI 7** Wavefunctions for a hexagonal partial dislocation network at several energies. Note that once again the wavefunction “segments” onto the partial network which is, as in the case of the experimental network, seen most clearly for states near the Dirac point. (a) States very close to the Dirac point localize on the AA vertices of the network (e = -0.006 eV), (b) higher energies (e = 0.025 eV) result in states localized on the grains of the individual AA and AB patches, while at energies distant from the Dirac point (c) e = 0.140 eV (d) e = -0.248 eV, the wavefunction has an additional structure resulting from the larger Bloch momentum of these states.