Atomic-scale transport in epitaxial graphene


The high carrier mobility of graphene\(^1\)-\(^4\) is key to its applications, and understanding the factors that limit mobility is essential for future devices. Yet, despite significant progress, mobilities in excess of the 2 \(\times 10^5\) cm\(^2\) V\(^{-1}\) s\(^{-1}\) demonstrated in free-standing graphene films\(^5\)-\(^6\) have not been duplicated in conventional graphene devices fabricated on substrates. Understanding the origins of this degradation is perhaps the main challenge facing graphene device research. Experiments that probe carrier scattering in devices are often indirect\(^7\), relying on the predictions of a specific model for scattering, such as random charged impurities in the substrate\(^8\)-\(^10\). Here, we describe model-independent, atomic-scale transport measurements that show that scattering at two key defects—surface steps and changes in layer thickness—seriously degrades transport in epitaxial graphene films on SiC. These measurements demonstrate the strong impact of atomic-scale substrate features on graphene performance.

Our results are based on scanning tunnelling potentiometry to measure local electric potential as current flows through a graphene film. By measuring local perturbations caused by substrate steps and changes in graphene thickness, we demonstrate that such heterogeneity is critical to transport in graphene. Substrate steps alone can increase the resistivity several-fold relative to a perfect terrace, and direct calculation shows that resistance arising from the intrinsic wavefunction mismatch will always exist at junctions between monolayer and bilayer graphene. The performance of graphene devices on SiC surfaces is thus fundamentally limited by the ability to control both the layer thickness and substrate perfection.

Figure 1a and b show low-energy electron microscopy (LEEM) images obtained immediately after graphene growth on two SiC(0001) substrates with different step densities (see Methods). The graphene thickness can be determined straightforwardly with LEEM from the reflectivity of the low-energy electrons, which depends on thickness through quantum confinement effects\(^11\). In Fig. 1a, the sample consists of \(\sim 80\%\) monolayer graphene (grey regions labelled 1), \(\sim 10\%\) ‘buffer’ layer that has a C-rich 6\(_3\)\(\times\)6\(_3\) structure (white regions) and \(\sim 10\%\) bilayer graphene (dark regions). In Fig. 1b the fractions of monolayer and bilayer graphene (labelled 1 and 2 respectively) are almost equal. Identifying by atomic-resolution scanning tunnelling microscopy (STM) the same areas that were imaged by LEEM enables us to characterize monolayer, bilayer and buffer-layer graphene. Consistent with other work\(^12\)-\(^14\), each has a distinctive appearance (Fig. 1c,d). Because substrate and graphene step configurations can be determined through STM height measurements, we can obtain a comprehensive picture of nanoscale topography.

In Fig. 1e, we show the experimental approach used to obtain maps of the electrical potential through scanning tunnelling potentiometry\(^15\),\(^16\). Two static probes (1 and 3) contact the surface at a separation of \(\sim 500\) \(\mu\)m. A voltage applied between these probes induces current flow through the graphene sheet, while a third, scanning probe (2) measures the local potential.

The potential can be measured on the macroscale, by stepping probe 2 across the surface, or on the microscale, by scanning probe 2 over a smaller area, in which case the topography of the sample can be acquired simultaneously. (Note that traditional four-probe measurements require all probes to contact the surface.) Figure 1f shows the macroscale potential. The total resistance (including the contact resistance and the resistance of the graphene sheet) and total current passing through the graphene are also measured. The potential distribution in this two-dimensional system is then modelled\(^16\) as a Laplace problem with fixed boundary conditions at the tips (see Methods). By fitting the potential acquired along the line between probes 1 and 3, macroscopic conductivities \(\sigma_{\text{avg}}\) can be determined for the two samples (Table 1).

In Fig. 2, we show microscale potential measurements over regions measuring hundreds of nanometres. The topography and graphene thicknesses are shown in Fig. 2a. Without applying a voltage between probes 1 and 3, the potential map (Fig. 2d) is almost featureless, as expected. But when a voltage is applied (Fig. 2b,c), the maps show two distinct features: dramatic potential jumps at the step edges, and a potential gradient on the terraces. These effects change sign when the applied voltage is reversed (Fig. 2c,f), showing that the measurement is directly related to transport.

The terrace gradient demonstrates that graphene terraces have a finite conductivity, presumably due to random scattering sources at the terraces (such as defects\(^13\), long-range scattering\(^2\)-\(^10\) or phonons\(^8\)) and at the interface\(^13\), but the potential discontinuity at the step edges indicates additional scattering at these locations. Carrier scattering seems to be particularly strong at the heterogeneous junctions between monolayer and bilayer graphene, and weaker but still visible at locations where a uniform graphene bilayer crosses a substrate step (top right corner of each map). Potential profiles across two terraces and a monolayer–bilayer junction are shown for a series of applied voltages in Fig. 2i,j. The linear relationships between the terrace gradient and monolayer–bilayer jump and the applied voltage are demonstrated in Fig. 2k.

On the terraces, the linear dependence of slope on applied voltage suggests that the terraces are behaving Ohmically; the local electric field \(E\) (the potential change per unit length) is related to the local current density \(j\) by \(j = \sigma E\), where \(\sigma\) is a constant (but local) terrace conductivity. We cannot measure the current density locally. However, we can estimate it by noting that all measurements were made approximately half way between the fixed probes, where the average current density can be calculated from the measured total current using the Laplace equation above. The local current density may differ somewhat from the average current density, but this approach enables us to estimate local terrace conductivities for monolayer graphene (Table 1).

(Bilayer graphene shows similar resistivity to monolayer graphene, as can be inferred for example from Fig. 2i,j.) On the basis of
scanning tunnelling spectroscopy results also obtained on these samples (Supplementary Fig. S1) and angle-resolved photoemission spectroscopy measurements from the literature\cite{18,19}, we estimate the electron density in the monolayer graphene to be $\sim 10^{13}$ cm$^{-2}$ and the local mobility for monolayer graphene on terraces at 72 K to be $\sim 3,000$ cm$^2$ V$^{-1}$ s$^{-1}$ in both samples.

The monolayer–bilayer graphene junction also obeys Ohm’s law (Fig. 2k): the linear dependence of voltage jump $\Delta V$ on applied voltage and hence local electric field indicates that $\Delta V$ is also proportional to the local current density, that is, $\Delta V \propto j$. With the local current density $j$ estimated as above, the monolayer–bilayer junction resistance $\rho_{\text{step}}$ can be extracted (Fig. 3d) using $V = j \rho_{\text{step}}$.

Where a single, continuous layer of graphene crosses a substrate step, the effect is weaker but still quantifiable. In Fig. 3a,b, single-layer graphene crosses 0.5-nm-high substrate steps. Although the potential discontinuity at the steps is hard to discern in individual scan lines (Fig. 3b), averaging shows the magnitude of the effect (Fig. 3c). It is also clear from Fig. 3c that higher steps show a greater potential jump (for similar terrace gradient and hence local current density). Ohm’s law is followed (Supplementary Fig. S2), so values for the step resistance $\rho_{\text{step}}$ can be calculated. Although changes in graphene conductance near steps have been described\cite{20}, this technique provides a quantitative measure of the extra resistance.

Figure 3d summarizes the results for monolayer graphene crossing substrate steps, and for monolayer–bilayer junctions of different configurations. Monolayer graphene crossing single (0.5 nm) substrate steps shows a resistance of 6.9 $\pm$ 2.9 $\Omega$ m. The resistance seems to increase linearly with step height, 14.9 $\pm$ 3.6 $\Omega$ m for

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**Table 1 | Conductivity of graphene on SiC substrates with different miscut angles.**

<table>
<thead>
<tr>
<th></th>
<th>Low miscut ($\sim 0.06^\circ$)</th>
<th>High miscut (0.5$^\circ$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average conductivity $\sigma_{\text{avg}}$ from macroscale measurement (mS)</td>
<td>4.32 $\pm$ 0.09</td>
<td>1.46 $\pm$ 0.03</td>
</tr>
<tr>
<td>Conductivity of monolayer graphene $\sigma_t$ from microscale measurement (mS)</td>
<td>5.0 $\pm$ 0.7</td>
<td>4.4 $\pm$ 0.7</td>
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Figure 2 | Scanning tunnelling potentiometry of terraces and monolayer–bilayer junctions. a–f, Topography (a–c) and potential maps (d–f) recorded simultaneously (a.c. voltage $V_{ac} = 2$ mV, $I = 50$ pA; room temperature) with $V_{13} = 0$ (a,d), $V_{13} = 1.53$ V and total current 5.73 mA (b,e) and $V_{13} = -1.53$ V (c,f). The voltage range is shown on the colour scale; zero is arbitrary. The step heights and graphene thickness (labels in a and c) are identified from STM. The local current density midway between the fixed probes is $4 \times 10^{-6}$ A µm$^{-1}$. Simulated potential maps calculated using the experimental boundary potential conditions. Step edges $s_1$, $s_2$, and $s_3$ have resistances of 41, 13, and 69 Ω µm respectively. i,j, Line profiles of the potential averaged from the rectangle in b, shown for the $V_{13}$ values indicated (mV). Data are offset vertically for clarity. The terrace slopes on the monolayer and bilayer sides are similar for each voltage, suggesting that the resistivity of the bilayer graphene is similar to that of the monolayer graphene in this region. k, The electric field on the terraces (slopes in i and j; monolayer and bilayer terraces being similar) and the potential jump at the monolayer–bilayer junction (jump heights in i and j) as a function of $V_{13}$.

We find that the upper graphene layer is continuous over the monolayer–bilayer junction (Supplementary Fig. S1b), consistent with other reports$^{14}$. Given the continuous nature of the graphene sheet, the substantial resistance of the junction is perhaps counter-intuitive, and might suggest the presence of defects or scatterers at the graphene edge. To understand this, we calculated the resistance

1.0 nm steps and 24.7 ± 4.3 Ω µm for 1.5 nm steps. One example of bilayer graphene crossing a step is also included, and it follows the same trend. Monolayer–bilayer junctions have higher resistance, 20.9 ± 5.7 Ω µm and 28.4 ± 7.0 Ω µm for planar and stepped junctions respectively. Monolayer–bilayer junctions at a double-height step provide the highest resistance seen here, 88 Ω µm.
using a standard tight-binding model of the monolayer and bilayer wavefunctions. For energies near the Dirac point, a continuum approximation is generally considered adequate. However, in view of the considerable doping here, we use a full atomistic approach with exact boundary conditions (effectively equivalent to the non-equilibrium Green function approach used for overlapping nanoribbons; see Supplementary Information for details). The model has five parameters: in-plane and interlayer matrix elements $t$, bilayer bandgap $\Delta$ and the respective doping levels $E_g$, all of which are known for epitaxial graphene on SiC (refs 19,21). With these values ($t_{\text{in-plane}} = 3.1$ eV, $t_{\text{interlayer}} = 0.4$ eV, $E_g = 0.45$ eV in monolayer, $0.3$ eV in bilayer, $\Delta = 0.15$ eV), we calculate resistances $\sim 25 \mu$m and $\sim 45 \Omega \mu$m for junctions with armchair and zigzag orientation, respectively. The junction measured has predominantly armchair orientation. The agreement between calculated and measured values is striking, although this could be partly fortuitous given the uncertainties in doping level and bandgap. These results clearly show that a high resistance is an intrinsic property of an ideal monolayer–bilayer junction.

The $25 \Omega \mu$m resistance corresponds to an average transmission factor of $T \approx \frac{1}{2}$, consistent with that found at low doping. We find that the poor transmission is largely a result of the wavefunction mismatch between monolayer and bilayer, unavoidable because the bilayer wavefunctions have large amplitude on both layers: wavefunction matching requires intermixing with an evanescent state from a higher-energy band of the bilayer. Thus, wavefunction mismatch is an inherent characteristic of this interface, and calculations using standard methods confirm that this inherent mismatch is sufficient to account for the magnitude of resistance observed experimentally. We also find considerable interband (K–K) scattering for the armchair orientation, so chirality is not conserved. Even for the zigzag orientation, where by symmetry there is no K–K scattering, there is still a strong wavefunction mismatch; and the calculations suggest a large resistance, although the conservation of chirality for the zigzag orientation contributes to the lower resistance when compared with the armchair orientation.

For a continuous graphene layer going over a substrate step, we suggest that the origin of the step-induced resistance may be intrinsic, induced for example by $\sigma$–$\pi$ hybridization arising from the curvature of the graphene sheet near the top and bottom of the step. The combination of potential mapping and modelling has thus shown that substrate steps, terraces and thickness changes all contribute to the resistance. Each element can be treated as following Ohm’s law at the nanoscale. Steps and junctions strongly affect transport. A 0.5 nm substrate step contributes extra resistance equivalent to a $\sim 40$-nm-wide terrace. 1.0- and 1.5-nm-high substrate steps contribute resistance equivalent to $\sim 80$ and $\sim 120$ nm of terrace respectively, and monolayer–bilayer junctions contribute $\sim 100$ nm or more. We can verify this understanding of current flow by simulating the potential distribution across the sample. In Fig. 2g,h, we show a simulation with fixed boundary potentials taken from the experimental data in Fig. 2a–f. The results are in close agreement with the data, enabling resistances to be estimated that are consistent with the values obtained above from analysis of line profiles.
Macroscopic conductivity measurements thus provide only a small part of the full picture required to understand transport through graphene on SiC. Miscut steps, islands formed during the growth process and thickness variations will all reduce the macroscopic conductivity. For example, the two samples in Table 1 show similar (within 1%) local conductivities measured on single terraces. Yet in the high-miscut sample the macroscopic conductivity was $\sim 3x$ lower than the local value, whereas the low-miscut sample showed a difference of only $\sim 1.2$. Steps from even a 0.7° miscut should double the resistance in the miscut axis, we obtain the total current

$$I = \int_{-\infty}^{+\infty} -\frac{d}{dx} V(y,z) \left(\frac{1}{(d/2)^2 + y^2} + \frac{1}{(d/2)^2 + z^2}\right) dy \sigma_{\text{avg}} = 2\pi A \sigma_{\text{avg}}$$

where $\sigma_{\text{avg}}$ is the ‘macroscopic’ conductivity and $j$ is a fitting constant with units of voltage.

Finally, the macroscopic current density at the origin is given by

$$j = \frac{4\pi \sigma_{\text{avg}}}{d} = 2I \frac{d}{\pi d}$$

We measure $V$ in several places by stepping probe 2 along the line between probes 1 and 3. Fitting the $V$ values to equation (1) yields a value for $A$. We also measure $d$ and $l$ directly. From $A$ and $l$, we obtain the values for $\sigma_{\text{avg}}$ and $j$ used in the analysis. In reality, the contact tips have finite size and irregular shape, within which the potential is constant. The voltage difference between the two contacts is $V_{\text{rec}}$. To fit equation (1) we assume circular contacts of diameter $\sim 60\mu m$ estimated from scanning electron microscopy.

Received 11 July 2011; accepted 13 October 2011; published online 20 November 2011

References


**Acknowledgements**

We thank A. Ellis and M. C. Reuter of IBM for their assistance with experimental aspects of this work, and R. Möller and X. Chen for discussions.

**Author contributions**

S-H.J. carried out scanning tunnelling potentiometry experiments, J.B.H. and R.M.T. grew the graphene and carried out LEEM; J.T. and V.P. carried out the calculations; S-H.J., F.M.R., J.B.H. and R.M.T. collaborated on equipment and experimental design; all authors wrote the paper.

**Additional information**

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturematerials. Reprints and permissions information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to S-H.J. or F.M.R.
Atomic Scale Transport in Epitaxial Graphene

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Electronic and atomic structure of graphene.
We use scanning tunneling spectroscopy to characterize the electronic structure of the graphene. The differential conductance, \(\frac{dI}{dV}\), measured by the lock-in technique, is related to the density of states of the graphene. The averaged \(\frac{dI}{dV}\) on the bilayer graphene shows a dip around -0.3V, Fig. S1(a), suggesting\(^1\) the Dirac point \(E_D\) level is -0.3eV. This indicates the sample is electron doped, consistent with angle-resolved photoemission results\(^2,3\). For the monolayer graphene, the \(E_D\) level can not be clearly resolved by \(\frac{dI}{dV}\) measurement, but the doping level of \(n \approx 10^{13} \text{cm}^{-2}\) can be estimated from Refs. 2 and 3. High resolution STM imaging around the monolayer-bilayer junction shows a continuous lattice across the boundary, Fig. S1(b).

Figure S1 Electronic and atomic structure of graphene. a, The density of states measured on bilayer graphene. Results are similar on the 0.5° and 0.06° samples. b, STM image of the monolayer-bilayer graphene junction (12nm×12nm, \(V=50\text{mV}, I=0.1\text{nA}\)). Monolayer graphene (1) shows the honeycomb structure while bilayer graphene (2) shows a hexagonal lattice due to the AB stacking. The continuity of the top graphene layer is visible. Inset is a schematic of the monolayer-bilayer graphene junction with buffer layer omitted for clarity.
Potential profiles of monolayer graphene crossing steps.

Fig S2 indicates the Ohmic behavior for monolayer graphene crossing substrate steps of different heights.

![Figure S2](image)

**Figure S2** Potential line profiles recorded for monolayer graphene across substrate steps. a, Step height 0.5nm; b, step height 1.5nm. The *V*\(_{13}\) values are indicated. The lines in a indicate the terrace gradient to highlight the small but measurable potential drop at the step.

**Calculation of transmission through a bilayer/monolayer junction.**

To find the transmission coefficient through a monolayer-bilayer junction we solve the scattering problem within a tight-binding approximation. The Hamiltonian equation \( H |\Psi\rangle = E |\Psi\rangle \) describes the bilayer for atoms with coordinate \( x \leq 0 \) and the monolayer for \( x > 0 \), according to Eqs. (36) and (5) of Ref. 4, respectively. We use the same notations as in Ref. 4, with tight-binding parameters \( t = t_{\text{in-plane}} = 3.1 \text{ eV}, t' = 0, \gamma_b = \gamma_{\text{interlayer}} = 0.4 \text{ eV and } \gamma_s = 0 \). In the scattering problem the component of wavevector parallel to the junction is conserved. The incoming wave is described by a wavevector whose component \( k_0 \) normal to the junction has \( \text{Im}(k_0) = 0 \) and positive group velocity \( dE/dk_0 > 0 \).

**Zig-zag edge bilayer/monolayer junction:**

For the zig-zag edge K and K’ states are not mixed, because a line with a constant \( k_y \) can not cross both K and K’ points in the extended Brillouin zone. Therefore, there can only be two reflected \( x \)-components \( k_1 \) and \( k_2 \) in the bilayer, describing a wave in the first subband \( \text{Im}(k_1) = 0 \) with negative group velocity \( dE/dk_1 < 0 \) and an evanescent wave (when the energy \( E \) is below the second subband as in our case) with \( \text{Im}(k_2) > 0 \), respectively. Note that \( k_1 \neq k_0 \) because of trigonal warping. In the monolayer a transmitted wave with a positive group velocity has wavevector \((k_3, k_y)\), where \( k_3 \) is determined by having the same energy and \( k_y \) as the incident wave, and by the condition \( \text{Im}(k_3) = 0 \) and \( dE/dk_3 > 0 \). The general solution for the wavefunction has the form

\[
\Psi = \Psi_1 R_1 + \Psi_2 R_2 + \Psi_3 R_3 + \Psi_4 R_4
\]
\[
\left( \begin{array}{c}
\Psi_{b1}(\vec{R}) \\
\Psi_{a1}(\vec{R}) \\
\Psi_{a2}(\vec{R}) \\
\Psi_{b2}(\vec{R})
\end{array} \right) = \left( \begin{array}{c}
b_1(k_0,k_y) \\
a_1(k_0,k_y) \\
a_2(k_0,k_y) \\
b_2(k_0,k_y)
\end{array} \right) \exp\left( i(k_0R_x + k_yR_y) \right) + \sum_{i=1,2} r_i \left( \begin{array}{c}
b_i(k_y,k_y) \\
a_i(k_y,k_y) \\
a_2(k_y,k_y) \\
b_2(k_y,k_y)
\end{array} \right) \exp\left( i(k_yR_x + k_yR_y) \right)
\]

(1a)

for \( x \leq 0 \), and

\[
\left( \begin{array}{c}
\Psi_A(\vec{R}) \\
\Psi_B(\vec{R})
\end{array} \right) = t_3 \left( \begin{array}{c}
a(k_3,k_y) \\
b(k_3,k_y)
\end{array} \right) \exp\left( i(k_yR_x + k_yR_y) \right)
\]

(1b)

for \( x > 0 \).

Here \( \vec{R} \) is a primitive unit cell coordinate\(^4\) and for every atom in the bilayer, the wavefunction at that atom is given by Eq. (1a), and for every atom in the monolayer, by Eq. (1b). The momentum-dependent coefficients \((b_1, a_1, a_2, b_2)\) and \((a, b)\) are fixed by satisfying the Hamiltonian equation \( H |\Psi\rangle = E |\Psi\rangle \) for the bilayer and monolayer away from the boundary, as explained in Ref. 4. Applying the operator \( H-E \) to a wavefunction of the form Eq. (1) then gives zero contribution except at the interface, where there are terms depending on the scattering amplitudes \( t_{1-3} \). These amplitudes are found by requiring that these remaining interface contributions vanish, so that \( H |\Psi\rangle - E |\Psi\rangle = 0 \) everywhere, including the atoms at the boundary. Note that a similar approach applies for an incoming wave from a monolayer to bilayer, and results in an identical transmission coefficient.

**Armchair edge bilayer/monolayer junction:**

For the armchair edge, the propagation direction is along \( y \) and the conserved wavevector is \( k_y \). The K and K′ states are mixed now, because a constant \( k_y \) line can cross both K and K′ points. The general solution for the wavefunction has the form:

\[
\left( \begin{array}{c}
\Psi_{b1}(\vec{R}) \\
\Psi_{a1}(\vec{R}) \\
\Psi_{a2}(\vec{R}) \\
\Psi_{b2}(\vec{R})
\end{array} \right) = \left( \begin{array}{c}
b_1(k_y,k_0) \\
a_1(k_y,k_0) \\
a_2(k_y,k_0) \\
b_2(k_y,k_0)
\end{array} \right) \exp\left( i(k_yR_x + k_0R_y) \right) + \sum_{i=1,4} r_i \left( \begin{array}{c}
b_i(k_y,k_y) \\
a_i(k_y,k_y) \\
a_2(k_y,k_y) \\
b_2(k_y,k_y)
\end{array} \right) \exp\left( i(k_yR_x + k_yR_y) \right)
\]

(2a)

for \( y \leq 0 \), and

\[
\left( \begin{array}{c}
\Psi_A(\vec{R}) \\
\Psi_B(\vec{R})
\end{array} \right) = \sum_{j=5,6} r_j \left( \begin{array}{c}
a(k_y,k_j) \\
b(k_y,k_j)
\end{array} \right) \exp\left( i(k_yR_x + k_yR_y) \right)
\]

(2b)

for \( y > 0 \).

Here for every atom in the bilayer, the wavefunction at that atom with coordinate \( y \leq 0 \) is given by Eq. (2a), and for every atom in monolayer with \( y > 0 \) by Eq. (2b). The incoming
wave is described by the wavevector \((k_x, k_0)\) with \(\text{Im}(k_0)=0\) and positive group velocity \(dE/dk_0>0\), and reflected waves in the first subband at \(K\) and \(K'\) points have negative group velocities \(dE/dk_{1,2}<0\) with \(\text{Im}(k_{1,2})=0\) and two evanescent modes in the second subbands at \(K\) and \(K'\) with \(\text{Im}(k_{3,4})>0\), correspondingly. In the monolayer, the two transmitted waves with a positive group velocity have wavevectors \((k_x, k_5)\) and \((k_x, k_6)\), where \(k_{5,6}\) are determined by having the same energy and \(k_x\) as the incident wave, and by the condition: \(\text{Im}(k_{3,6})=0\), \(dE/dk_{5,6}>0\). Applying the operator \(H-E\) to a wavefunction of the form Eq. (2) then gives zero contribution except at the interface, where there are terms depending on the scattering amplitudes \(r_{i=1-6}\). These amplitudes are found by requiring that these remaining interface contributions vanish, so that \(H \Psi - E \Psi = 0\) everywhere. Note that a similar approach applies for scattering for an incoming wave from a monolayer to bilayer, and results in an identical transmission coefficient.

**References**