The development of the micromechanical cleavage technique for manufacturing ultra-thin graphitic films has made it possible to produce FETs based upon monolayer graphene and to study their electronic properties, indicating that graphene is a gapless semiconductor with a peculiar Dirac-type spectrum of charge carriers. Using micromechanical exfoliation, multilayers of graphene are also often produced. Two layers of graphene prepared by exfoliation usually exhibit crystalline ordering with a characteristic AB stacking, later referred to as single-crystal (SC) bilayer. This should be contrasted to pairs/stacks of individual misoriented graphene flakes identified among some of the peeled graphitic films using Raman spectroscopy and in multilayers grown epitaxially on Si$_3$N$_4$. While the individual carrier density in such layers is usually beyond experimental control, in this paper we report on the realisation of two-layer graphene-based FETs where the density on the two monolayers can be varied separately, despite them lying only a dozen Angstroms apart.

The nanostructures presented in this letter were prepared by peeling off pieces from natural bulk graphite with adhesive tape and placing them on a silicon wafer covered with SiO$_2$. Since, at the intermediate stage of the utilized deposition process only some part of the graphene flake touches the SiO$_2$ surface, the flake flips over during the removal of the adhesive tape, thus producing two misoriented graphene layers lying on top of each other and separated by occasional surface deposits (Fig. 1a sketches the top-view of such a structure).

Then, this material is processed into Hall-bar samples using plasma etching and contacted (to both layers) by evaporating chromium and gold electrodes (Fig. 1b). Figure 2a shows two optical images of such a sample before and after plasma etching. A darker than other areas top-left edge indicates the position of the fold. Scanning the structure with an atomic force microscope (AFM) reveals a double step at the etched edges (Fig. 2c and d) with a first-step height of 1.1 nm. Although layers of graphene should have a height of 0.34 nm, previous AFM measurements showed values up to 1 nm for single layer graphene lying on a substrate attributed to a water layer underneath graphene and/or the rippling graphene surface.

From these data and also from the transport measurements shown below we conclude that the first step is due to a monolayer of graphene. The second step has a height of 0.6 nm which is larger than the thickness of one layer but less than the height of two AB stacked layers. This indicates that the second layer is also a single layer which is separated further from the first layer than in a conventional bilayer.

To distinguish the designed device from monolayers or SC bilayers we performed transport measurements on the sample shown in Fig. 2, applying magnetic fields up to 13 T at temperatures down to 1.5 K. To reduce the influence of contact resistance on the measurements, a multi-terminal device has been used, and different combinations of two and three point measurements were performed at a broad range of back-gate voltage, $-70 \text{ V} < V_{BG} < 70 \text{ V}$, applied between the substrate of n-Si and graphene. As usual, positive back-gate voltage induces electrons, negative holes. Figure 2b shows the measured field effect on graphene resistivity demonstrating the operation of the device as an FET. A character-

![FIG. 1: a) Top view of a pair of arbitrarily oriented graphene monolayers (Layer 2, red and Layer 1, blue) in a two-layer stack. b) A two-layer stack on top of a Si/SiO$_2$ (grey/light grey) wafer, etched and contacted (yellow) for transport measurements.](image-url)
imate neutrality condition for the graphene layers, which is shifted to a finite back-gate voltage $V_{BG}^0 = 11.5$ V indicating natural doping of the graphene flakes. The measured maximum resistivity, $\rho_{max} \approx 3.3$ kΩ at 1.5 K, is about half of the earlier-reported typical values for monolayers\textsuperscript{2,4}, which is in line with the assumption that the device consists of two monolayers conducting in parallel.

Most importantly, the layer structure of ultrathin graphitic films can be characterized by analyzing the Shubnikov-de Haas (SdH) oscillations in the flake resistance $R(B)$ studied as a function of a magnetic field $B$ applied perpendicular to the sample. The Berry’s phase $\pi$ characteristic of electrons in monolayer graphene is directly related to the appearance of SdH oscillations minima at filling factors $\nu_{min} = 4(i + 1/2)\pm \pm 1$, in contrast to SC bilayer graphene\textsuperscript{5,6} where the electron’s Berry’s phase is $2\pi$ and the SdH oscillations minima appear at filling factors $\nu_{min} = 4i$, with $\nu = nh/eB$ being the filling factor, $n$ the carrier concentration, and $i$ an integer. When studied at a fixed back-gate voltage (e.g. $V_{BG} = -70$ V and $V_{BG} = -40$ V, Fig. 3a,b), the magneto-resistance measured in the two-layer device and plotted versus $1/B$ displays oscillations with two very different periods (compared in detail in the top/bottom panels of Fig. 3 a,b) manifesting the co-existence of two very different carrier densities, $n_1$ and $n_2$ in the sample. These two density values can be obtained from the period $\Delta B^{-1}$ of the SdH oscillations, as $n = 4e/\hbar \Delta B^{-1}$ (where we take into account both the spin and valley degeneracy in the graphene band structure). Their experimentally determined gate-voltage dependence is shown in Fig. 3d. Systematically repeated at all the studied voltage values, the periodicity of both fast (lower panel) and slow (upper panel) $1/B$-oscillations in the two-layer structure was such that the resistivity minima could only be attributed to the sequence of filling factors $\nu_{min} = 4(i + 1/2)$, which is typical for monolayer graphene.

To highlight this behavior of the presented data, in the top/bottom panels of Fig. 3a,b the $1/B$ intervals with the rising parts of the corresponding oscillations are colored in red/blue. For comparison, Fig. 3c shows the results of a similar study performed for a SC bilayer, where the sequencing of the SdH oscillations minima coincides with the earlier observed\textsuperscript{2} appearance of maxima at filling factors $\nu_{min} = 4i$ specific for such a material. Finally, the observed carrier densities $n_1$ and $n_2$ in the two-layer device and their dependence on back-gate voltage presented in Fig. 3d can be compared to theoretically calculated values plotted on the same figure as black lines. The presented calculation takes into account the electrostatics of the device containing two monolayers at distance $d \approx 1.5$ nm from each other lying on top of the SiO\textsubscript{2} dielectric layer (with permittivity $\varepsilon_0 = 3.9$ and thickness $L_{BG}$). Fermi-energy dependence of the compressibility of the electron gas with the Dirac spectrum $E(p) = vp$, and additional doping charge, $\delta n \approx 2 \times 10^{11}$ cm$^{-2}$ due to deposits left on top of the upper layer in the course of the manufacturing process.
The parameters used in this calculation were obtained by fitting the observed $V_{BG}$-dependence with the analytical result relating the two densities on two parallel graphene flakes,

$$2\sqrt{\frac{\pi d(e^2/\epsilon_0)}{n_2 + \delta n}} = \frac{\hbar v}{s_1 \sqrt{|n_1| - s_2 \sqrt{|n_2|}}}$$

$$n_1 + n_2 + \delta n = \frac{e_0 e V_{BG}}{e L_{BG}}$$

where the first equation states the equivalence of the electro-chemical potentials on the two graphene layers with $s_i = n_i/|n_1|$, whereas the second relates the total charge density in the device to its electrostatic capacity.

To tune the two carrier densities independently on the two parallel layers, we fabricated a two-layer sample with an additional top gate. A schematic view of the manufactured device is shown in Fig. 4a, with the design resembling that of the top-gated SC bilayer structures used recently. To fabricate the top gate, polymethylmethacrylat (PMMA) was spun onto a Hall-bar device which was produced in a similar way as the sample shown in Fig. 2. The PMMA layer was partially exposed to an electron beam converting the PMMA into a layer insoluble by acetone. Putting the sample in an acetone bath leaves an insulating PMMA layer of about 60 nm on top of the graphene flake. A local top gate with an area of 2.5 $\mu$m$^2$ was fabricated on top of this PMMA layer using standard electron beam lithography. Figure 4b shows an image of the top-gated sample. The resistance measured in this device as a function of the back-gate voltage for a fixed top-gate voltage, $V_{TG}$, is shown in Fig. 4c (for $V_{TG} = 15$ V). Its back-gate voltage dependence contains two pronounced peaks (marked by arrows) indicating the difference between the carrier concentrations in the top-gated and free parts of the device. The back-gate voltage interval between the two peaks corresponds to the bipolar transistor regime, with a p-n-p junction formed underneath the top gate. The tunability of the device using the top gate is demonstrated in Fig. 4d. Here, the back-gate voltage was kept fixed at $V_{BG} = -40$ V and the top-gate voltage, $V_{TG}$, was varied. Due to the large negative back-gate voltage, the resistance maximum corresponding to the 'neutral point' in the top-gated region appears at positive voltage $V_{TG} = 26$ V (not shown in the figure). To improve the visibility of the Shubnikov-de Haas oscillations and their evolution with $V_{TG}$, we differentiate the device’s resistance with respect to the magnetic field value and show the result using the blue-scale plot in Fig. 4d where one sees several sets of oscillations. The oscillations which are almost independent of the top-gate voltage originate from the areas outside the top-gated region. The oscillations with a strong $V_{TG}$ dependence characterize the layers underneath the top gate. The deconvolution of these two contributions is achieved by the numerical subtraction of the $V_{TG}$ independent part of the resistance. The resulting magnetoresistance are shown in Figs. 4e,f as function of filling factor for $V_{TG} = -14$ V and $V_{TG} = -31$ V. Similarly to the first two-layer device, the top-gated structure shows the superposition of two monolayer-type ShdH oscillations with different periods corresponding to densities $n_2 = 6.32 \cdot 10^{12}$ cm$^{-2}$ and $n_1 = 1.08 \cdot 10^{12}$ cm$^{-2}$ for $V_{TG} = -14$ V and $V_{BG} = -40$ V, and to $n_2 = 7.44 \cdot 10^{12}$ cm$^{-2}$ and $n_1 = 1.80 \cdot 10^{12}$ cm$^{-2}$ for $V_{TG} = -31$ V and $V_{BG} = -40$ V. This demonstrates not only that the different carrier densities on the two closely laid graphene layers can be detected, but also that those densities can be independently controlled and tuned using a combination of top/backgates.
In conclusion, we demonstrated the realization of a graphene-based field-effect transistor containing two decoupled graphene monolayers at a nanometer distance from each other. Using the magnetotransport measurements, we determine the carrier densities on the two parallel layers and show the ability to control those densities separately using a combination of electrostatic (back & top) gates. When operated in a broad voltage range, such a device could be employed in a search for the recently predicted superfluidity\textsuperscript{14} in the 'excitonic insulator' state expected to form in a pair of graphene layers with opposite polarity. Also, the technique of layer folding offers a promising method for making devices with separately contacted pairs of monolayers acting as atomically-thin optically-transparent\textsuperscript{15,16} electrodes to study vertical transport and electro-optical characteristics of nanoparticles of various materials that can be captured between them.

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