Electromechanical oscillations in bilayer graphene

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Nanoelectromechanical systems constitute a class of devices lying at the interface between fundamental research and technological applications. Realizing nanoelectromechanical devices based on novel materials such as graphene allows studying their mechanical and electromechanical characteristics at the nanoscale and addressing fundamental questions such as electron-phonon interaction and bandgap engineering. In this work, we realize electromechanical devices using single and bilayer graphene and probe the interplay between their mechanical and electrical properties. We show that the deflection of monolayer graphene nanoribbons results in a linear increase in their electrical resistance. Surprisingly, we observe oscillations in the electromechanical response of bilayer graphene. The proposed theoretical model suggests that these oscillations arise from quantum mechanical interference in the transition region induced by sliding of individual graphene layers with respect to each other. Our work shows that bilayer graphene conceals unexpectedly rich and novel physics with promising potential in applications based on nanoelectromechanical systems.

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Separating graphene from the substrate by suspension permitted investigating its intrinsic electronic properties and allowed unveiling ultrahigh electron mobilities due to reduced scattering and the observation of the fractional quantum Hall effect. Similarly, suspension allowed incorporating graphene into nanoelectromechanical systems (NEMSs) to fabricate resonators and opened the way to explore the interplay between the mechanical and the electronic properties of graphene. However, during suspension, nanoscopic ripples form to ensure the thermodynamic stability of the two-dimensional crystal, and strain is inevitably introduced in suspended graphene. There is experimental evidence that strain strongly affects the physical properties of graphene. As the crystalline symmetry is broken under strain, it induces lifting of the twofold degeneracy of the optical phonon vibrational modes. This is observed as a splitting in the G peak in Raman spectra of strained graphene. Very high pseudo-magnetic fields, reaching up to 300 T, have been predicted and confirmed experimentally in highly strained graphene nanobubbles and increased electrical resistance, reaching 5% under 3% tensile loads, has been reported for graphene nanoribbons (GNRs) with widths ranging between 0.8 and 1.2 μm.

On the other hand, bilayer graphene has been reported to show appealing physical properties such as the possibility of opening and tuning an electronic bandgap or engineering of quantum dots for a single-electron manipulation. These properties make bilayer graphene emerge as a complementary material to single-layer graphene and open the way towards all-carbon-based circuits where graphene could be used as high-mobility conductor, while bilayer graphene could in principle ensure electronic functionalities such as modulation or switching. The effect of strain on electrical transport in bilayer graphene has been investigated only theoretically. Here we integrate mono- and bilayer graphene into NEMS to study the effect of strain on their transport properties. Mono- and bilayer GNRs with widths between 60 and 300 nm are investigated using nanoindentation techniques based on atomic force microscopy (AFM) for high-resolution imaging and controlled deformation of the GNRs. Electrical conductance of the suspended GNR is measured simultaneously with mechanical deformation using a low-noise lock-in amplifier. While monolayer graphene displays increasing resistance with strain, we observe oscillations in the electromechanical response of bilayer graphene. This behaviour is explained from the point of view of quantum interference in the transition region induced by sliding of individual graphene layers with respect to each other.

**Results**

**Device fabrication and description.** Our samples consist of field-effect transistors based on suspended GNRs (Fig. 1a). The devices were fabricated using a combination of standard electron beam lithography lift-off processes with oxygen plasma etching to define the nanoribbon geometry, followed by hydrofluoric acid wet etching and critical point drying. Initial electrical characterization is performed with voltage sources connected in the configuration depicted in Fig. 1b. We apply a bias to the source electrodes and a back-gate voltage to the degenerately doped silicon substrate. The drain current is measured using a current–voltage converter connected to a lock-in amplifier. The transfer and output characteristics of a monolayer- and a bilayer-suspended device are presented in Supplementary Fig. 1. All our devices have a linear characteristic typical of ohmic contacts. The gating dependence shows ambipolar dependence with the charge neutrality point lying at the conductivity minimum and corresponding to the Dirac point.

**Electromechanical measurement set-up.** Following the electrical characterization, we place the sample under AFM for imaging and nanoindentation, with Supplementary Fig. 2 showing typical AFM images of our devices. The imaging conditions are discussed in Supplementary Note 1. Before the nanoindentation experiment, the photodetector sensitivity and the AFM cantilever spring constant were calibrated (Supplementary Fig. 3), while the determination of the AFM geometry (Supplementary Fig. 4) and its resonance frequency (Supplementary Fig. 5) complete the calibration (Supplementary Note 2). Once the suspended GNR is located and the set-up is calibrated, the AFM tip is positioned on top of the GNR for indentation. By moving the stage upwards against the cantilever and downwards far away from it, the GNR is deformed and then relaxed. This cycle of extension–retraction of the stage is represented in Fig. 2a. During each deformation cycle, we simultaneously measure the current (upper graph) and the cantilever deflection, as a function of the position of the stage, as shown in Supplementary Fig. 2. We extract the deformation of the GNR ribbon at the point where load is applied by the AFM tip from the expression (ref. 19). Detailed finite element modelling (FEM) has shown that the deflection of underetched contact areas (Fig. 1b) can be neglected (Supplementary Figs 6 and 7; Supplementary Note 3). During the experiment, an a.c. bias voltage with a root mean squared amplitude of 4 mV and a frequency of 8 kHz is applied to the GNR and the current flowing through it is monitored using a lock-in amplifier.

![Figure 1](image-url)  
**Figure 1 | Device and experimental set-up.** (a) Scanning electron microscope (SEM) image of the device. A 60-nm-wide graphene nanoribbon is suspended above a substrate and contacted by electrodes. Scale bar, 500-nm long. (b) Schematic drawing of the experimental set-up and geometry. The suspended graphene ribbon is deformed in the centre using an AFM probe attached to a piezo scanner. The vertical displacement of the scanner results in the deflection of the cantilever and nanoribbon deflection. The device is biased by an a.c. voltage with a root mean squared amplitude of 4 mV. The resulting drain current is monitored using a lock-in amplifier.
Electromechanical response of monolayer graphene. Figure 2a shows the electromechanical response of monolayer graphene. The electrical and mechanical responses are reproducible for both the extension and the retraction cycles. From the mechanical point of view, this proves that we are deforming the GNRs in the elastic regime, that no structural defects are introduced in the extension and the retraction cycles. From the mechanical origin, ensuring that the observed current variations are of electro-mechanical origin.

Conductance oscillations in bilayer graphene. We now turn to bilayer graphene devices. We have performed electromechanical measurements on nanoribbons fabricated from bilayer graphene with widths of 200 and 300 nm and resistances of ~50 and ~40 kΩ, respectively. The response of these two devices is represented in Fig. 3. Similarly to the samples of monolayer graphene, the mechanical response of bilayer devices is reversible, reflecting the elastic regime of deformation and the mechanical stability of the suspended bilayer GNRs. We can see from measurements of device current \( I_d \) as a function of piezo scanner extension, Fig. 3a, that the electromechanical response of bilayer GNRs shows two main features. Similarly to monolayer GNRs, we see an overall increase of resistance as a function of nanoribbon deformation. During the approach cycle and before mechanically contacting the GNRs the current is constant. The current undergoes variations only once the GNR is deformed ensuring that the observed current variations are of electromechanical origin. In the case of monolayer graphene, we have performed electromechanical measurements on five devices with widths varying between 60 and 300 nm (Supplementary Fig. 2; Supplementary Table 1), resistances ranging between 10 and 100 kΩ with device mobilities \( \sim 1,500 \text{ cm}^2\text{V}^{-1}\text{s}^{-1} \). We observe a decrease of the current under strain for both natural (device #1) and chemical vapour deposition (CVD) graphene (device #2 to device #5). We observe no opening of a bandgap in this deformation regime.

To compare the electromechanical response of the various GNRs, we represent the relative variation of the resistance \( \Delta R/R_0 \) as a function of nanoribbon deflection \( D_{\text{GNR}} \) (Fig. 2b). In almost all the devices (devices 1–4), we see an increase of electrical resistance as a result of strain \( \varepsilon \), with a positive piezoresistive gauge factor (GF) defined as \( GF = (\Delta R/R_0)/\varepsilon \). This observed strain-induced resistance increase is in agreement with the previous reports on monolayer graphene and is related to decreasing Fermi velocity and reduced mobility. Interestingly, one of our devices (device #5) shows a decrease in resistance as a function of strain, corresponding to a negative gauge factor. This rare behaviour indicates that additional effects, possibly dependent on the lattice orientation, could modulate the electrical behaviour of graphene under strain and warrants further theoretical modelling.

Because the sharp AFM tip with a radius of \( \sim 30 \text{ nm} \) (Supplementary Fig. 4; Supplementary Note 2) introduces local strain in the nanoribbon centre, resulting in non-uniform strain in the nanoribbon (Supplementary Fig. 8; Supplementary Note 4), we use FEM to determine strain distribution in the ribbon, caused by the indenting AFM tip. Results show that, although in the vicinity of the AFM tip the dominant contribution to strain is from the local deformation under the sharp tip, in the rest of the ribbon, the strain is almost uniform and due to the overall vertical deflection of the ribbon (Supplementary Fig. 9; Supplementary Note 4). This allows us to estimate the upper limit on the gauge factor by taking into account the uniform strain induced by the vertical deflection while neglecting the contribution localized around the AFM tip. For devices 1–4, showing a positive gauge factor, we find an upper limit for GF of 8.8, in good agreement with previously published results and at least 20 times lower than in semiconducting atomically thin layers of MoS\(_2\) (ref. 23; a more detailed discussion is available in Supplementary Note 4).

The same simulation results show that the highest achieved strains in our GNRs are \( \sim 5\% \) (Supplementary Fig. 10; Supplementary Note 5).
The overall, background increase of resistance in bilayer devices can be explained in terms of the piezoresistive response, just as in the case of monolayer devices. The gauge factor of bilayer devices is, however, lower than that of monolayer devices. Calculations by Wong et al.\textsuperscript{24} show that tensile strain can increase interlayer interactions in bilayer graphene, which could partially compensate decreasing intralayer interactions, resulting in a smaller gauge factor for bilayer graphene.

**Theoretical simulations of charge-carrier transport.** Clearly, interlayer interactions are at the origin of the striking observation of electromechanical oscillations. We propose a simple theoretical model capable of quantitatively reproducing the observed behaviour. The model assumes that the AFM tip action causes finite lateral displacement (sliding) of the individual graphene layers with respect to each other. This lateral displacement is expected due to the weak van der Waals force between the two graphene layers.\textsuperscript{25,26} There is extensive evidence in the literature that lateral displacement of graphene layers with respect to each other takes place in various types of scanning probe microscopies.\textsuperscript{27–36} The relative displacement of a single graphene layer on another graphene layer is even more likely when accompanied by deformation of the layers.\textsuperscript{37} In our experiment, the AFM tip deforms the bilayer GNRs, which leads to an increase in the elastic energy of the system. Therefore, the necessity for lowering the energy of the system results in the relative displacement of the layers. The lateral displacement, however, disturbs the AB stacking of the two layers, which is the energetically preferred configuration of bilayer graphene.\textsuperscript{38} The system then reduces the interlayer binding energy through formation of a ‘domain wall’-like transition region separating two AB-stacked domains, as described by the Frenkel–Kontorova model\textsuperscript{39} (Fig. 4a). The transition region essentially accommodates the transition to the incommensurate phase and in turn allows the AB stacking in the two regions of GNR on each side of the ‘domain wall’ that would appear as a localized bulge, or wrinkle, for the reasons discussed below.\textsuperscript{38,39} The width of these transition regions, typically a few nanometres according to experimental observations\textsuperscript{39} and our numerical estimates (Supplementary Note 7), is defined by the balance between the total strain energy and the interlayer binding energy.\textsuperscript{39,40} Such boundaries occurring between AB- and BA-stacked regions have recently been observed in misoriented multilayer graphene by numerous groups.\textsuperscript{26,29,33,35,36,38} Displacement of stacking domain boundaries and manipulations and creation of wrinkles in scanning tunnelling microscopy was demonstrated\textsuperscript{27,32,34,36}. Displacement and removal of wrinkles with an AFM tip were also shown experimentally.\textsuperscript{28,30,31} The details of this process were investigated by means of realistic simulations.\textsuperscript{41}
We note, however, that the effective width of the transition region within the two individual graphene layers measured on the graphene lattice, \( W_1 \) and \( W_2 \), is different (see Fig. 4a for definition). Moreover, this effective width difference \( \Delta W = W_1 - W_2 \) will vary as the two graphene layers slide against each other due to the action of the AFM tip. If no transition region was initially present in the sample, \( \Delta W \) corresponds to the lateral displacement of one graphene layer with respect to another on indentation. In the transition region, the layers are electronically decoupled either due to their incommensurate stacking\(^{42} \) for small values of \( \Delta W \), or due to enlarged interlayer distance for larger \( \Delta W \) giving rise to a wrinkle (localized bulge) in one of the layers of bilayer graphene, as illustrated in Fig. 4a. We suggest that the observed electromechanical oscillations can be explained from the point of view of quantum interference phenomena due to path difference \( \Delta W \) of the charge carriers in the decoupled graphene layers. Increasing the strain leads to higher amount of local corrugation and changes \( \Delta W \), which would cause constructive or destructive interference.

To verify this hypothesis, we perform numerical simulations of electronic transport in a model bilayer graphene device (Supplementary Fig. 11). The simulations are performed in the ballistic regime because of the few nanometres width of the transition region. The methodology is based on a tight-binding Hamiltonian and non-equilibrium Green’s function formalism (see Supplementary Note 8 for details). Without loss of generality, the individual graphene layers are assumed to be fully decoupled (zero interlayer hopping integrals) in the transition region. Our model device is a bilayer graphene with the zigzag direction aligned to the transport direction, and is periodic in the perpendicular (armchair) direction. We investigated armchair direction of domain boundaries as this orientation was found to be dominant for the case of closely related AB–BA stacking domain boundaries extensively investigated using transmission electron microscopy\(^{38} \). We assume that the sample is large enough that edge effects do not affect significantly transport properties. Furthermore, considering the in-plane isotropic elasticity of graphene, the crystallographic orientation of the transport channel does not affect the electromechanical behaviour. Figure 4b shows the calculated charge-carrier transmission probabilities as a function of energy \( E \) and momentum parallel to the transition region, \( k_\parallel \), for various carrier path differences \( \Delta W = na \) \((n \in \mathbb{Z}), a = 0.246\text{ nm}\)the lattice constant of graphene\). The trivial case of \( \Delta W = 0 \) (no transition region, that is, pristine bilayer graphene) reveals the massive character of Dirac fermions in bilayer graphene. Finite path differences \( \Delta W \) result in significant amount of backscattering developing a clear sub-band sequence resulting from the quantum confinement of massless Dirac fermions in the transition region (indicated by the dashed line in the last panel of Fig. 4b). Most importantly, configurations characterized by \( \Delta W = 3ma \) \((m \in \mathbb{Z})\) show enhanced transmission due to constructive interference as the wavenumber of Dirac fermions in graphene \( k = 4\pi/(3a) \). Therefore, one period of oscillations corresponds to the deformation-induced lateral displacement of \( 3a = 0.74\text{ nm} \).

To gain further insight, we compare the calculated resistance with actual experimental observation. A quantitative comparison requires accounting for the role of contacts as well as for the diffusive transport in the rest of device. Both factors, below collected in a single value \( R_C \), act as a ‘bottleneck’ in a realistic device and are thus responsible for most of its total resistance. We obtained \( R_C = 41\text{ k\Omega} \) by fitting both the average value of the calculated resistance and the magnitude of oscillations to the experimental data reported in Fig. 3b for device \#7. In Fig. 4c, oscillations in resistance can be seen clearly with a constant period proportional to \( 3a \). The direct comparison of Figs 3b and 4c reveals a quantitative agreement with experiments, except for
Methods

Device fabrication. Single- and bilayer sheets of graphene have been exfoliated from commercially available crystals of graphite (NGS Naturgraphite GmbH) using the Scotch-tape micromechanical cleavage technique. CVD graphene sheets have been obtained by growth of graphene on a 99.8% pure, annealed, copper foil (Alfa Aesar) following a two-step growth regime. The graphene is then transferred to top of a Si substrate covered with a 270-nm-thick SiO2 layer. The surface of the samples is imaged using an optical microscope (Olympus BX51 M) equipped with a colour camera (AVT Pike F-905C). Suspended nanoribbons are prepared using two steps of electron beam lithography. In the first step, the ribbons shape is defined using a polymethyl methacrylate (PMMA) mask and exposure to O2 plasma, followed by mask removal in acetone. The electron beam lithography step was used to define contacts using a standard bilayer technique (methyl methacrylate (MMA)/PMMA) followed by metallization and lift-off in acetone. The graphene channel is released and suspended using hydrofluoric acid wet etching and critical point drying.

Electromechanical characterization. Electrical characterization of the devices is carried out using Agilent E5270B parameter analyser and a home-built shielded probe station with micromanipulated probes. AFM imaging and electromechanical nanoindentation experiments are performed using a home-built set-up combining the Asylum Research Cypher AFM with the low-noise lock-in amplifier (SRS-830). We use nonconductive Si AFM tips, model NSC36/AlBS from MikroMash (Supplementary Note 2).

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Author contributions

M.M.B. fabricated the devices, performed the experiment and analysed the data. S.M. fabricated devices, performed the experiment on an additional set of devices and carried out FEM simulations. A.K. designed the experiment and supervised the project. F.G., G.A. and O.V.Y. performed theoretical calculations. M.T. performed CVD graphene growth. M.M.B., S.M., F.G., G.A., O.V.Y. and A.K. wrote the manuscript.

Additional information

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