Opening nm-sized gaps in graphene for molecular spintronics applications

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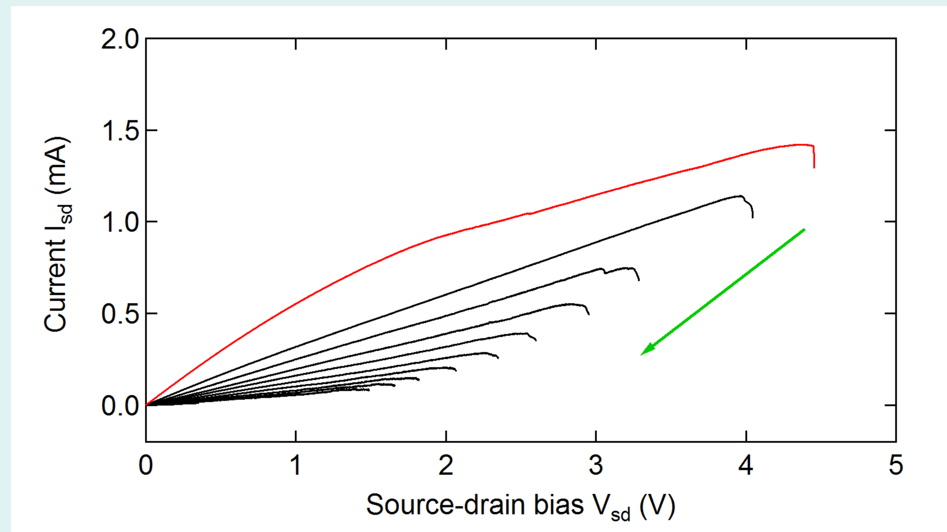
Abstract. Nanometer-spaced gaps in graphene are appealing candidates for various applications, ranging from advanced quantum devices to single-molecule electronics and even DNA sequencing. They can be obtained by applying an electroburning (EB) process, which exploits the chemical reaction of carbon atoms with oxygen at the high temperatures induced by Joule heating at large current densities. The introduction of a fast feedback loop avoids the abrupt breaking of the graphene junctions and allows for a more precise control on the final structure of the graphene nanogaps. Here, we present a systematic characterization of the feedback-controlled EB, showing that it is a versatile and reliable method to open nanometer-sized gaps with a high yield in different types of graphene devices and in different working conditions.

1. Feedback-controlled electroburning (EB)

Operating principle (F. Prins et al., Nano Lett. (2011) **11**, 4607):

- the bias voltage V_{sd} is swept across the graphene junction (at typical rates of 20-50 mV/s)
- the current I_{sd} is continuously measured, so that the variations of the junction resistance ($R = V_{sd}/I_{sd}$) can be monitored
- an increase of R by more than a predefined percentage triggers the *feedback* control, which sweeps the voltage back to zero in less than 10 ms
- the process is then repeated, thereby gradually narrowing the graphene junction
- the loop ends when the low-bias (V_{sd} = 200 mV) resistance exceeds a predefined threshold **[1]**



The 1st ramp is highlighted in red and the green arrow shows the evolution of the EB process

2a. Large-area graphene

Few-layer graphene grown on the C-face of SiC

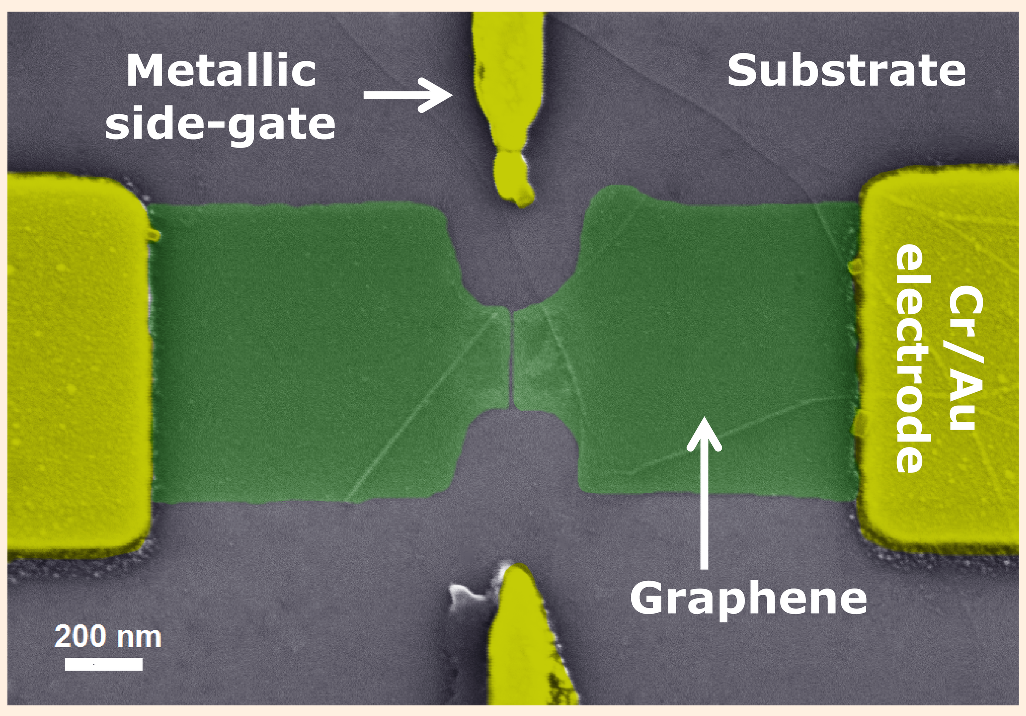
Threshold resistance: 100 kΩ – 5 MΩ

Yield of EB at room T in air: 87 %

CVD single-layer graphene transferred on a SiO_2/Si substrate

Threshold resistance: 1 MΩ – 50 MΩ

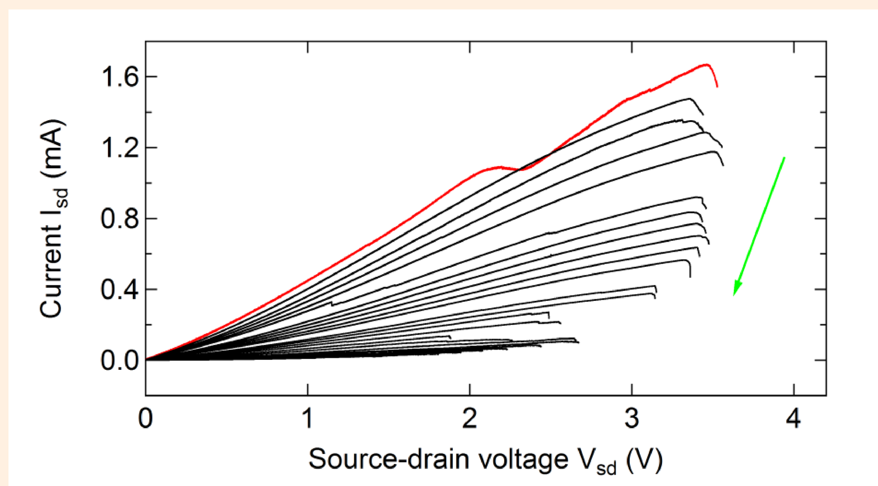
Yield:
- EB at room T in air: 75 %
- EB at room T in vacuum: 70 %
- EB at T = 20 mK in vacuum: 50 %

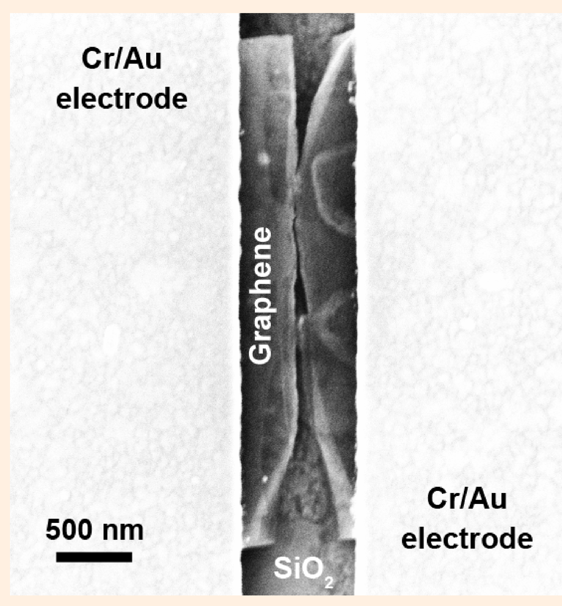


2b. Suspended graphene

Exfoliated few-layer graphene suspended over a SiO_2/Si substrate **[2]**

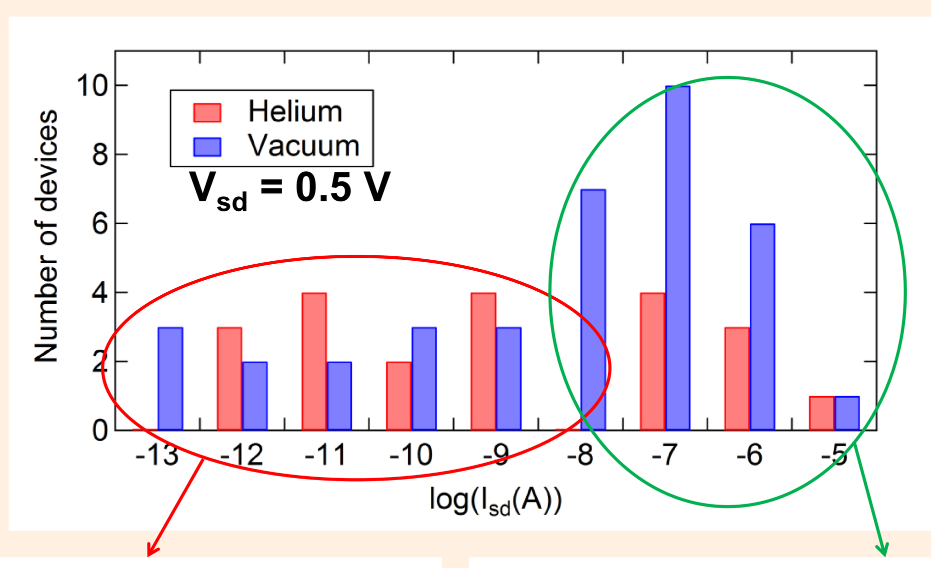
Yield:
- EB at room T in vacuum: 91 %
- EB at room T in ^{4}He: 81 %

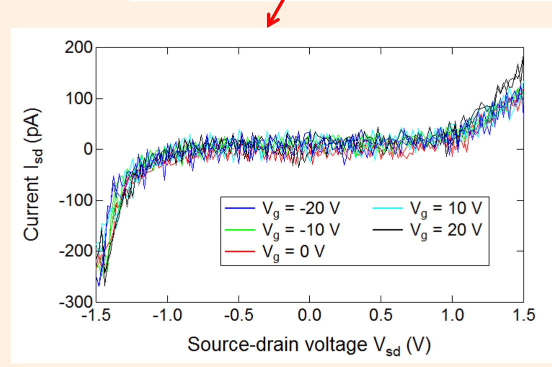


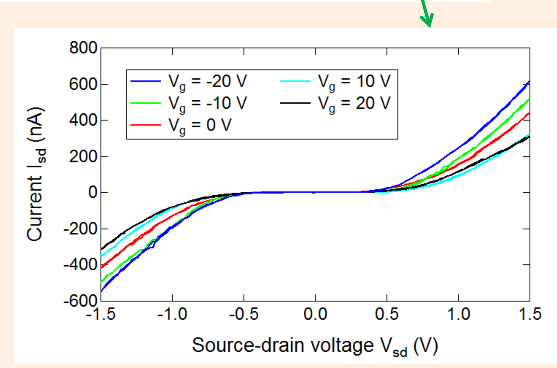


The backgate is kept floating during the EB process to avoid the electrostatically induced collapse of the graphene devices

After EB







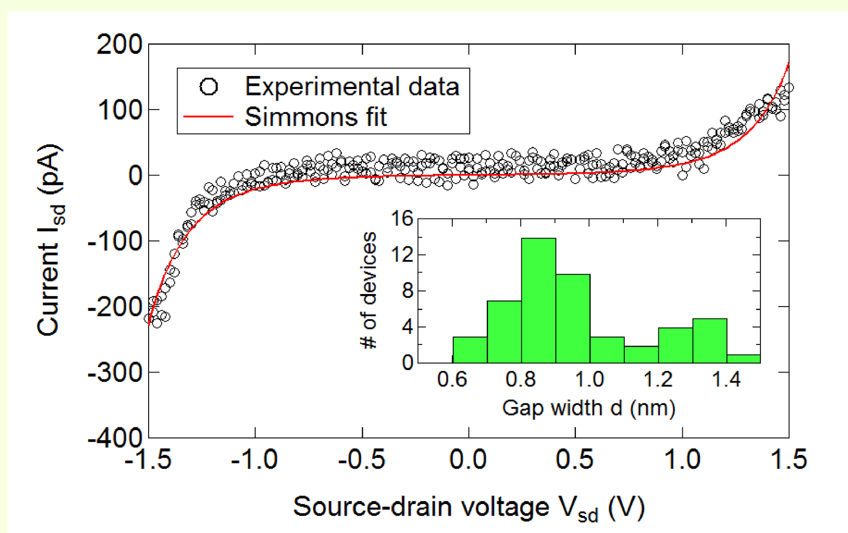
No dependence of the tunneling current on the gate voltage: the nanogap is completely formed

I-V curves strongly affected by the gate potential: residual connections left between the graphene electrodes

3. Gap size estimation using the Simmons model

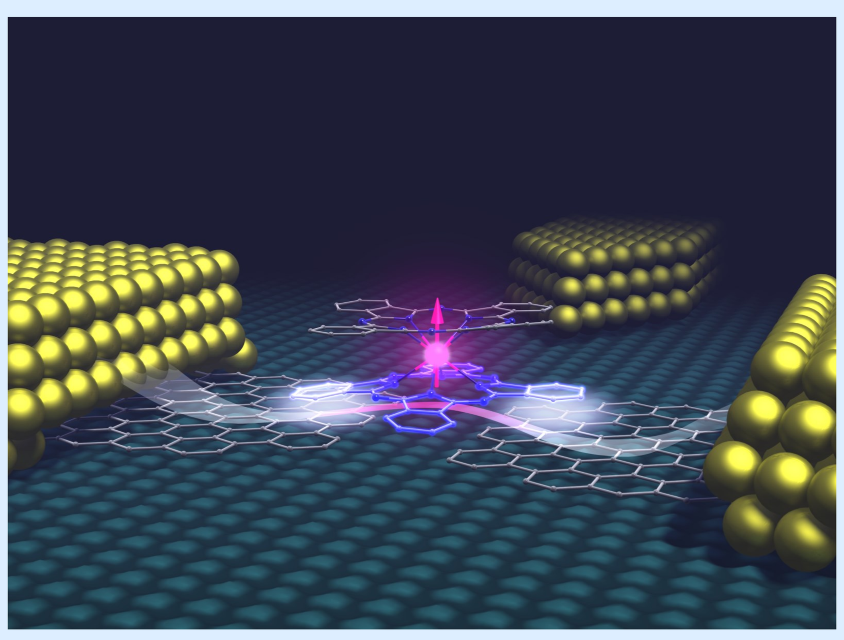
After EB, the I-V curves are fitted using the Simmons model (J. G. Simmons, *J. Appl. Phys.* (1963) **34**, 1793)

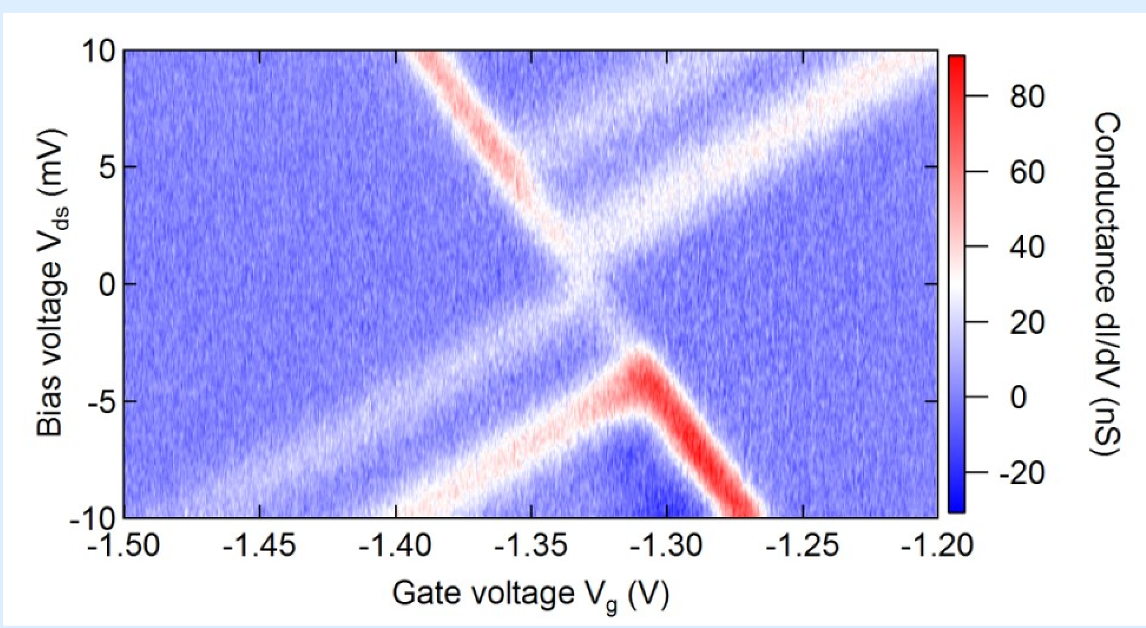
The gap size is estimated to be in the few-nanometer range, suitable for contacting single molecules: $d \lesssim 5\,nm$



4. Single-molecule spintronics

Devices with $TbPc_2$ molecules embedded within nano-gapped graphene electrodes work in the Coulomb blockade regime **[3]**:





References:

[1] A. Candini et al, *Beilstein Journal of Nanotechnology* **6**, 711 (2015)
[2] S. Lumetti et al, *Semiconductor Science and Technology* (2016) submitted
[3] S. Lumetti et al, *Dalton Transactions* **45**, 16570 (2016)