Supplementary information

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We provide here additional details on the electronic structure of the system in the setups discussed in the manuscript as well as on the interface between the graphene electrodes and the DB loop. We focus here on the local atomic structure of the electrode-DB contacts and their electronic DOS. We also shortly describe the recursive algorithm used to numerically solve the transport problem for larger loops.

Electronic structure issues– If the long and short edges of the ring are reversed with respect to the 2×1 periodicity of the surface lattice transmission function would be suppressed due to the weak coupling between dimer rows [F. Ample, I..Duchemin, M. Hliwa, and C. Joachim J. Phys.: Condens. Matter 23, 125303 (2011)]). To illustrate this we calculate transmission for the geometry shown on Fig. S1, where the loop consists of 14 DB, build perpendicularly to the Si dimer rows. One can see that around the Fermi energy transmission reaches the value of 10^{-3}, which is lower then transmission of the loops discussed in the main text (see Fig. 5 in the manuscript). However, we expect the similar interference effects just less pronounced due to the lower transmission values. In order to demonstrate role of direct electron tunneling though the substrate we calculate the transmission

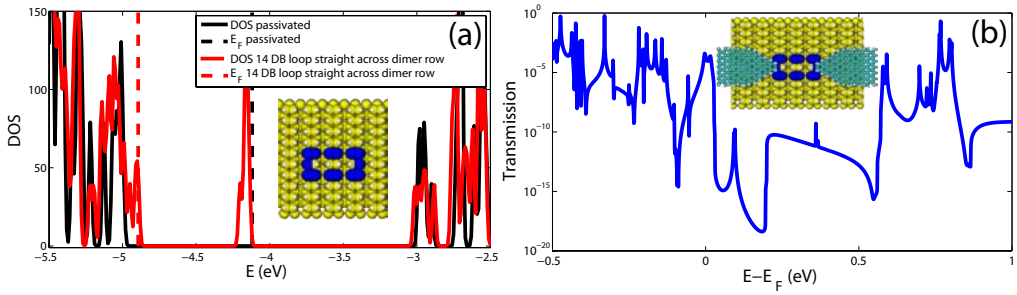

Fig. S1 Electronic density of states (DOS) of the loop built perpendicularly to the dimer rows (a). Also shown as reference is the corresponding DOS for the fully H-passivated Si(100) surface. Vertical dotted lines indicate the positions the Fermi level E_F in the different cases. The corresponding quantum mechanical transmission is shown in panel (b), where the inset illustrates the studied DB configuration. Notice the strong suppression of the transmission around the Fermi energy.

function of the system made with two DBs on different dimer rows,coupled to the graphene nanoribbon leads. In this case transport of electrons can occur only by direct tunneling perpendicularly to the dimer rows through the Si substrate. We compare it with the transmission function through the DB loop S2. One can see that around the Fermi energy transmission drops to 10^{-11}, which is much lower then in the both cases of destructive and constructive interferences. However around $E - E_F = 0.21$ the transmission peak, associated with the direct tunneling between two DBs, is present in both cases. Exactly only this type of peaks remain in the case of charge localization on the interface between leads and DB loop discussed above.

Multiple-contact topologies between graphene nanoribbons and DB atoms– The graphene nanoribbons were put on an H-passivated Si(100) surface at a distance of 3.5 $\AA$. The nanoribbon edges are also passivated with H atoms, with exception of the triangular region, which mediates the electronic contact to the DB loop atoms. During the relaxation process, which ensures the

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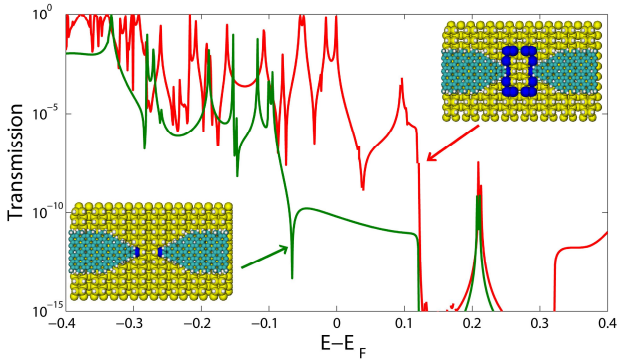

Fig. S2 Transmission function around the Fermi energy for transport within the DB loop (in red) and direct tunnelling between two DBs (in green).

maximal force component to be below 10^{-7} eV/Å, the electrodes bend towards the closest Si dangling bond atom establishing a connection of the system to the leads with a 1.92 Å Si-C bond distance. This bond length is in good agreement with the experimental and simulation data reported before, see for example [Xu, Y.; He, K.; Schmucker, S.; Guo, Z.; Koepke, J.; Wood, J.; Lyding, J.; Aluru, N. *Nano letters* **2011**, 11, 2735-2742]. The DOS for the different contact geometries described in the main text is shown in S3. We have considered a straight loop made of five DBs along the dimer rows and four in the perpendicular direction. In the case of the single-atom contact, case (a), the position of the Fermi energy is only slightly shifted to +0.04 eV with respect to the DB loop alone (see top panel of Fig. S3), while in the cases (b) and (c) the relative change of the Fermi energy amounts to +0.26 eV. Moreover, for (b) and (c) the DOS around the Fermi level contains mainly contributions from states localized at the interface between the leads and the Si DBs (b,c). These states are weakly coupled to the rest of the DB loop, having an effective gap of 0.28 eV. These properties of the electronic structure are also reflected in the transmission function and are responsible for the strong suppression of quantum transport at low energies. These results reveal the way of how the lead should be connected to the DBs. The resulting states should be a mixture of both coming from the leads and DBs or having a small energy gap between them, such that they will be close to the Fermi level.

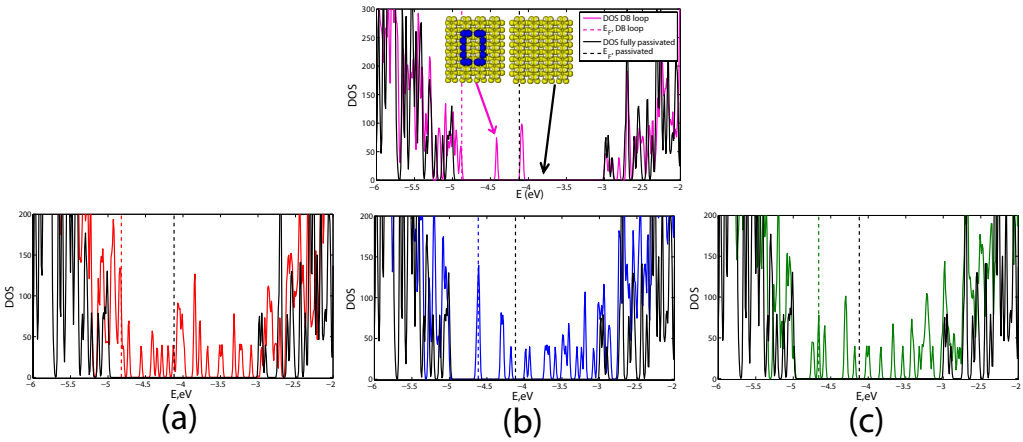

Fig. S3 Top panel:(DOS) of straight (magenta curve) DB loop. Also shown as reference the corresponding DOS for the fully H-passivated Si(100) surface. Bottom panel: DOS the DB loop coupled to the graphene nanoribbon electrodes that terminate with one (in red, (a), two (in blue, (b)) and three (in green, (c)) C atoms. In black is the DOS of the fully passivated Si. The dotted lines show the corresponding position of the Fermi energy.

Recursive Green's function techniques– Modeling of larger DB loops is not practically achievable within a full first-principle framework, so that we use recursive techniques, building the system slice by slice in the transport direction as it is shown on S4. We construct the Green's function separately for odd and even slices (between the dashed lines in S4) extracting the Hamiltonian and Overlap matrices from the first-principle calculations of the DB loop of smaller sizes. Here we used the assumption, supported by our DFTB calculations, that the Hamiltonian and Overlap matrices are approximately equal for each odd or even slice. This together with the computed self-energies $\Sigma^{r}_{\mathrm{L,R}}$ and self- energies of bottom and top parts of the system $\Sigma_{\mathrm{top,bottom}}$ allows us to calculate the transmission function between two leads fully taking into account the influence of the substrate as well as a varying number of slices N_y and loop geometry (straight or zigzag).

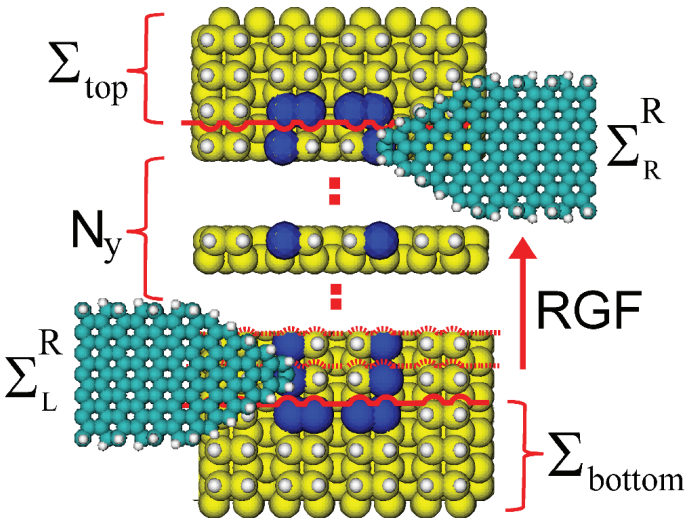

Fig. S4 Schematic illustration of the RGF procedure over N_y slices between two leads with self energies $\Sigma^{R}_{L,R}$. Each slice is defined as a part of the system between dashed lines, the solid line shows the part that contributes to the $\Sigma_{\mathrm{top,bottom}}$.

Conductance oscillations as a function of the loop length: The tight-binding model for the loop introduced in Fig. 6 of the mnauscript, does not allow for analytical results clarifying the beat-like behavior found in the transmission as a function of the loop length. This is related to the fact that (i) the system is non-periodic and (ii) the hopping integrals are different along the rows and between them, so that closed analytical expressions e.g. for the dispersion relation, are not possible to obtain. We have therefore considered a simplified version of the loop by considering a ladder model as shown in Fig. S5. We have here three different nearest-neighbor electronic couplings: t_1, t_2, and t_v. A loop configuration can be obtained, if the coupling $t_v \to 0$. If we first set $t_v = t_2$, but keep $t_1 \neq t_2$ and consider an infinite ladder, then the model can be solved analytically. One obtains

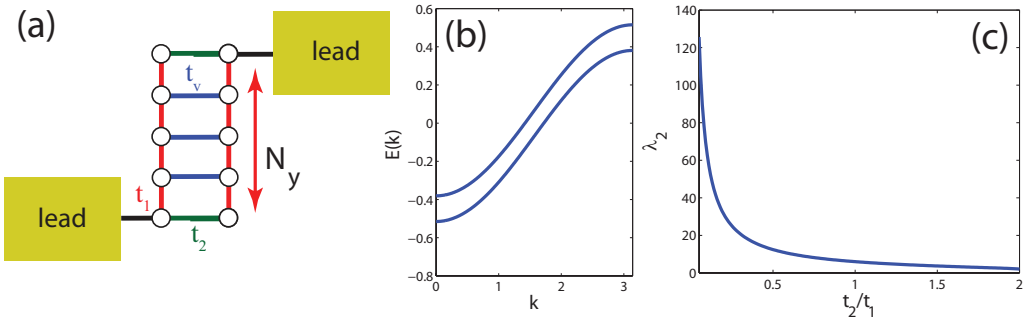

Fig. S5 (a) Simplified ladder model; (b) dispersion relation in the case $t_v = t_2$, but $t_1 \neq t_2$: $E = -2t_1\cos(k) \pm t_2$; (c) Dependence of λ_2, as defined by Eq. 2, on the ratio t_2/t_1 at $E = 0$.

two electronic bands with dispersion relations $E_{\pm} = -2t_1\cos(k) \pm t_2$. to which two different wave vectors can be associated for each given energy, see panel (b) of Fig. S5. This simple model displays a beat-like behavior in the transmission function as a function of the number of "stairs" in the ladder, related to two different length scales characterized by the wavelengths $\lambda_1 = \frac{2\pi}{k_1+k_2}$ and $\lambda_2 = \frac{2\pi}{|k_1-k_2|}$. Here, analytical expressions can be given for these two different length scales ($\lambda_{1,2}$ are given in units of the tight-binding lattice spacing a):

$$\lambda_1 = \frac{2\pi}{|\arccos\frac{-E+t_1}{2t_2} + \arccos\frac{-E-t_1}{2t_2}|} = 2 \quad (1)$$

$$\lambda_2(E) = \frac{2\pi}{|\arccos\frac{-E+t_1}{2t_2} - \arccos\frac{-E-t_1}{2t_2}|}. \quad (2)$$

As we can see from these expressions, while the short wavelength λ_1 is only determined by the lattice spacing, the second wave length $\lambda_2(E)$, giving the longer period of the oscillations, depends not only on the electronic couplings, but also on the energy E in a non-trivial way. This closed result only holds for $t_v = t_2$, but gives already a hint that no simple relation can be established between the long scale modulation of the transmission and the model parameters. If we now consider the original situation $t_v \neq t_2$ and progressively reduce the size of t_v to go to the limiting case of a loop configuration with only two hopping integrals, we also see a progressive increase of the long wavelength of the beating, as shown in Fig. S6. The values of λ_2 indicated as insets were now computed numerically, since Eq. 2 is not valid anymore for $t_v \neq t_2$.

It is clear that the previous discussion provides only an indirect way of rationalizing the beating found in our calculations for the dangling bond loops (both for the model presented in the manuscript as well as for the atomistic DBs), but since there is no analytical solution for the λ_1, λ_2 wavelengths in the most general case of the DB loops, our main aims have been (i) to illustrate the fact that there is no simple relation between incoming energy, the loop size and the long wavelength modulating the short-range odd-even oscillations in the transmission, and (ii) that the beat-like effect turns out to be quite robust when going from this simple model-based description to the more complex loop model and to the real DB system.

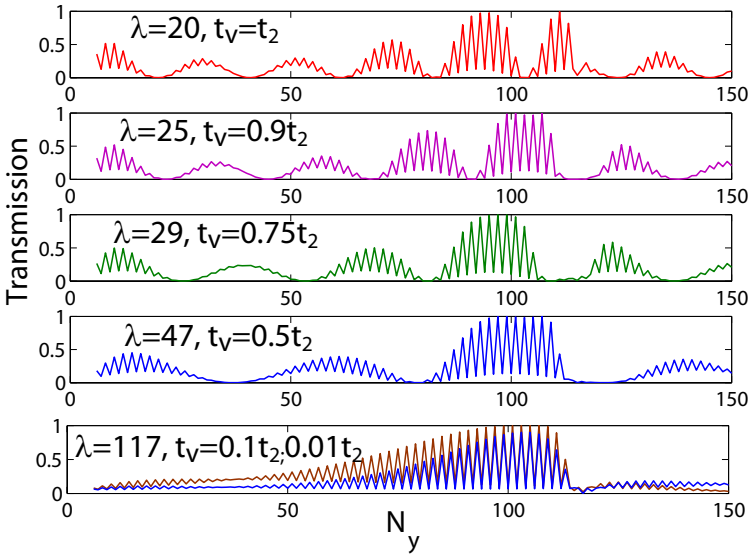

Fig. S6 Transmission function at a fixed energy $E = 0.89$ as a function of the number of sites N_y in the ladder model of Fig. S5, with $t_1 = 0.22$ eV, $t_2 = 0.3t_1$ and different values of the ratio $t_v/t_2 = 1; 0.9; 0.75; 0.5; 0.1; 0.01$. At $t_v = t_2$ the formula for λ_2 in Eq. 2 works well and gives 20.

Concerning now the short period odd-even oscillations, there is a general argumentation that has been used over the past years to explain the presence of odd-even oscillations in atomic chains and molecular wires: For not too strong coupling to the electrodes, the maxima of the transmission function are located at the position of the eigenstates of the decoupled (from the electrodes) system. In the case of odd N there is always a level in the spectrum exactly at the Fermi energy for symmetry reasons

leads to a maximum of the linear conductance. On the contrary, when N is even there is no level at the Fermi energy and therefore the system exhibits a lower conductance.

I-V characteristics. Fig. S7 shows the *I-V* characteristics of the DB loops with straight and zigzag topologies. The current is computed as $I(V) = \frac{2e}{h}\int_{-\infty}^{\infty} T(E)\,[f_{\mathrm{R}}(E) - f_{\mathrm{L}}(E)]\,dE$, here $T(E)$ is the zero bias transmission, and $f_{\mathrm{L(R)}}(E)$ are the left (right) electrode Fermi functions. As already predicted for the infinite wires, the current through the zigzag loop is considerably higher and sets in at lower bias ($\sim$ 0.05 V) when compared with the straight loop. The current mediated by the DB states dominates up to 0.2 V and 0.15 V for zigzag and straight DB loops, respectively; afterwards charge transport through the conduction band starts and the current leaks into the Si substrate.

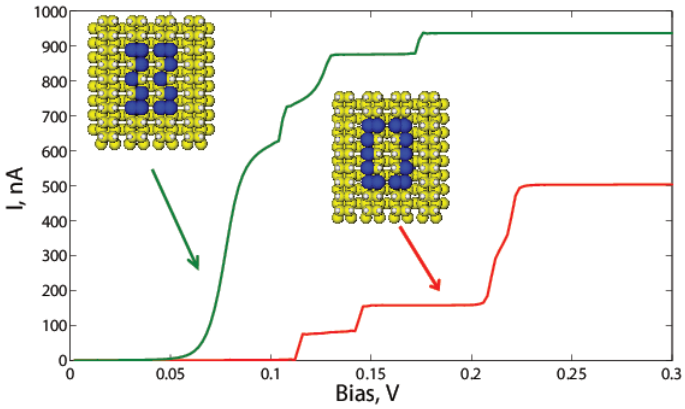

Fig. S7 Current as as function of bias voltage through the zigzag (green) and straight (red) DB loops, for a symmetric coupling of the graphene nanoribbons to the DB loop.

The interference effects are less pronounced in the current vs. loop length plots shown in S8, since by integrating we average the phases over the bias energy window. However for small voltages, when the DB loops surface states start to conduct, for both loop topologies the quantum interference effects are still noticeable.

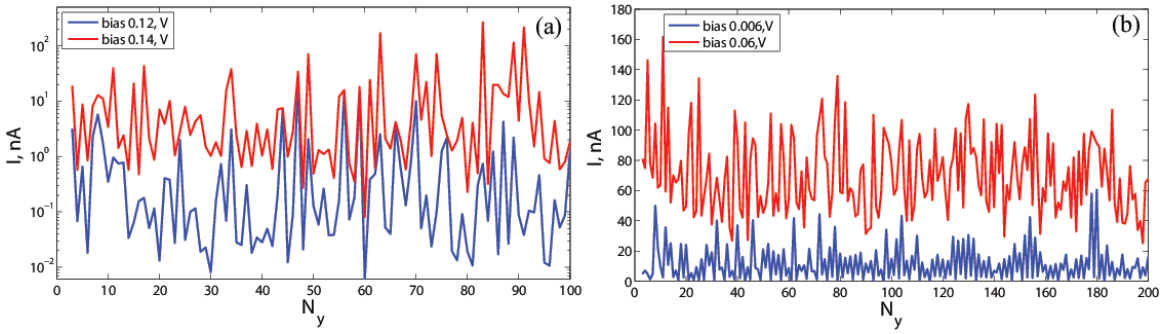

Fig. S8 Current as a function of the DB loop length N_y, calculated at bias 0.12 V and 0.14 V for the straight (a) and bias 0.006 V and 0.06 for zigzag (b) topology.