Electronic Excited States in Bilayer Graphene Double Quantum Dots

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We report tunneling spectroscopy experiments on a bilayer graphene double quantum dot device that can be tuned by all-graphene lateral gates. The diameter of the two quantum dots are around 50 nm and the constrictions acting as tunneling barriers are 30 nm in width. The double quantum dot features addition energies on the order of 20 meV. Charge stability diagrams allow us to study the tunable interdot coupling energy as well as the spectrum of the electronic excited states on a number of individual triple points over a large energy range. The obtained constant level spacing of 1.75 meV over a wide energy range is in good agreement with the expected single-particle energy spacing in bilayer graphene quantum dots. Finally, we investigate the evolution of the electronic excited states in a parallel magnetic field.

Keywords: graphene, bilayer graphene, quantum dot, double quantum dot, excited states

Graphene quantum dots (QDs) are interesting candidates for spin qubits with long coherence times \cite{1}. The suppressed hyperfine interaction and weak spin-orbit coupling \cite{2,3} make graphene and flat carbon structures in general, promising for future quantum information technology \cite{4}. Significant progress has been made recently in the fabrication and understanding of graphene quantum devices. A "paper-cutting" technique enables the fabrication of graphene nanoribbons \cite{5,13}, quantum dots \cite{14,19}, and double quantum dot devices \cite{20,22}, where a disorder-induced energy gap allows confinement of individual carriers in graphene. These devices allowed the experimental investigation of excited states \cite{16,21}, spin states \cite{19} and the electron-hole crossover \cite{17}. However, all of these studies were based on single-layer graphene and showed a number of device limitations related to the presence of disorder, vibrational excitations and to the fact that the missing band gap makes it difficult to realize soft confinement potentials and "well-behaving" tunneling barriers. In particular, it has been shown that intrinsic ripples and corrugations in single-layer graphene can lead to unintended vibrational degrees of freedom \cite{24} and to a coherent electron-vibron coupling in graphene QDs \cite{27}. Bilayer graphene is a promising candidate to overcome some of these limitations. In particular it allows to open a band gap by an out-of-plane electric field \cite{20,22}, which may enable a soft confinement potential and may reduce the influence of localized edge states. More importantly, it has been shown that ripples and substrate-induced disorder are reduced in bilayer graphene \cite{24}, which increases the mechanical stability and suppresses unwanted vibrational modes.

Here, we present a bilayer graphene double quantum dot (DQD) device, with a number of lateral gates. These local gates allow to tune transport from hole to electron dominated regimes and they enable to access different device configurations. We focus on the DQD configuration and show characteristic honeycomb-like charge stability diagrams, with addition energies on the order of 20 meV. Most interestingly, we observe electronic excited states, which are equally spaced over a wide energy range, in good agreement with the expected single-particle confinement energy in bilayer graphene QDs.

The DQD device shown in Figure 1a is fabricated based on bilayer graphene obtained from mechanical exfoliation of bulk graphite. The bilayer graphene flakes are deposited on a highly p-doped silicon substrate with a 295 nm silicon oxide layer. Raman spectroscopy measurements are used to unambiguously identify bilayer graphene. In Figure 1b we show the 2D Raman peak recorded on the flake which has been used to fabricate the device shown in Figure 1a. In contrast to single-layer graphene we find that four Lorentzians are required for fitting the 2D line shape. In particular, the left shoul-
The measurements have been performed in a pumped 4He system with a base temperature of 1.3 K and in a dilution refrigerator with an electron temperature around 100 mK. We have measured the two-terminal conductance through the bilayer graphene DQD by applying a symmetric dc bias voltage $V_b$ while measuring the current through the device with a resolution better than 50 fA.

The source-drain current of the device measured at finite bias ($V_b = 20$ mV) over a large BG voltage range is shown in Figure 1c. In close analogy to single-layer graphene nanodevices we observe a region of suppressed current separating the hole (left inset) from the electron transport regime (right inset). This so-called transport gap [8], which extends for the investigated device from roughly 32 V to 48 V is expected to be mainly caused by the local tunneling barriers formed by the three 30 nm narrow constrictions (see illustration in Figure 1d) [14, 15]. The reproducible sharp conductance resonances in and around the transport gap region are due to localized states in the constrictions whereas the overall hole-doping is most likely due to atmospheric O$_2$ binding [32].

In order to access the DQD regime we fix the BG voltage to a value inside the transport gap, such that the source and drain potentials are within the valence band (see arrow in Figure 1c and Fermi level depicted in Figure 1d). In Figure 2a we show a measurement of the current as function of the central gate voltage and the left and right gate voltages ($V_{ig^*} = V_{rg}$) while keeping the BG voltage fixed (arrow in Fig. 1c). Two characteristic slopes are observed which separate areas of suppressed current from the area of elevated current in the lower left corner. This measurement suggests that the energy diagram shown in Figure 1d (including a dominant central barrier) is a useful description for the three different transport regimes (1)-(3) found in the data. In regime (2) all three side gates are tuned to very negative voltages, such that all three effective tunneling barriers are pushed in the conduction band and hole transport takes place throughout the whole structure (c.f. Fig. 2b). By increasing the voltages on the left and right gate we enter regime (3), which leads to a significantly decreased current. In this configuration the Fermi level crosses the two tunneling barriers induced by the two outer constrictions (c.f. Fig. 2c). Thus the device is in a single quantum dot regime. Finally, if we increase the voltage on the central gate as well, the current is almost completely suppressed [region (1)]. The Fermi level crosses in this configuration all three effective energy gaps and the device is in the DQD regime (c.f. Fig. 1d). The LQD and RQD are consequently defined by the three energy gaps as shown in Figure 1d. The two slopes separating the different regimes are determined by the relative lever arms of the lateral gates (CG, LG* and RG) on the three tunneling barriers. While the two outer tunneling barriers can be controlled well by the left, right and the central gate ($\alpha = \Delta V_{ig^*} / V_{cg} \approx 0.55$), the dominating central barrier can only be weakly tuned by $V_{ig^*}$ and $V_{rg}$ ($\beta = \Delta V_{cg} / \Delta V_{ig^*} \approx 0.13$), which is in good agreement with the geometry of the device.

In Figure 2a we show a measurement of the current as function of the central gate voltage ($V_{cg}$) and the voltages applied to the outer side gates ($V_{ig^*} = V_{rg}$) at $V_b = 20$ mV and $T = 1.3$ K. (b,c) Schematic band structure depicting the pure hole transport regime (2) and a single quantum dot regime with lifted central barrier (3), respectively. The dashed lines denote the position of the Fermi level. (d) Charge stability diagram at finite bias ($V_b = 10$ mV) recorded in the double quantum dot regime (regime (1), see also Figure 1d) as highlighted by the black bar in panel (a). (e) Local resonances in the right conduction as function of the left and right gate voltages measured in regime (3) with additionally lifted left tunneling barrier ($V_{ig^*} = 0$ and $V_{lg} < 0$), such that only the right tunneling barrier crosses the Fermi level ($V_b = 10$ mV) [33].
and right gate voltages independently. In Figure 2d we show a finite bias ($V_b = 10$ mV, $T = 1.3$ K) measurement taken in regime (1) (see black bar in Fig. 2a). A honeycomb pattern characteristic for the charge stability diagram of a DQD can be observed [33]. Electrical transport through the DQD is only possible in the case where the energy levels in both dots are aligned within the source-drain bias window. In close analogy to single-layer graphene, the overall transmission through the DQD is modulated due to local resonances in the narrow constrictions. This can be seen best in Figure 2e, where we show a charge stability diagram recorded in regime (3) (c.f. Fig. 2c) but with lifted left barrier ($V_{lg} = 0$ and $V_{rg} < 0$). Thus the Fermi level only probes localized states in the right tunneling barrier. In this measurement we observe only one dominating slope ($\Delta V_{lg}/\Delta V_{rg} \approx 0.2$, dashed line), which moreover corresponds to the slope of the transmission modulation in Figure 2d [33]. In regions of high transmission (e.g. lower left corner in Fig. 2d) the connecting lines between the triple points become visible. Along these lines, only one of the dot levels is within the bias window, leading to current by cotunneling processes. The relative lever arms between the left and right gates acting on the two dots (LQD and RQD) are determined from the slopes of these cotunneling lines (see dashed lines in Fig. 2d) delimiting the hexagons, $\alpha_{lg,rg}^L = 0.85$ and $\alpha_{lg,rg}^R = 0.12$. Please note that in this system the RQD can almost equally be tuned with the RG as with the LG, which again is consistent with the device geometry.

A high-resolution close-up of Figure 2d, but with reversed bias, $V_b = -10$ mV is shown in Figure 3a. The bias-dependent extensions of the triangular-shaped regions allows the determination of the conversion factors between gate voltages and energies (see illustration in Fig. 3b) [33]. The lever arm between the left (right) gate and the left (right) dot is $\alpha_{lg}^L \cdot V_b/\delta V_{lg} = 0.056$ ($\alpha_{lg}^L = 0.019$) [33]. This allows to extract the single-dot addition energies $E_c^L = \alpha_{lg}^L \cdot \Delta V_{lg} = 23$ meV and $E_c^R = \alpha_{lg}^R \cdot \Delta V_{lg} = 13$ meV, which reflect an asymmetry in the QD island sizes. The two quantum dots are considered to be mainly formed by the width-modulated bilayer graphene structure and the local disorder potential. In particular, the disorder potential may have significant impact influencing the extend and location of the individual tunneling barriers [33]. In Figure 3c we show a similar measurement from a different cool down. The BG voltage has been fixed in the center of the transport gap. We observe slightly modified dot sizes with addition energies of $E_a^L = 18$ meV and $E_a^R = 21$ meV. Interestingly, the extracted addition energies are in reasonable agreement with values from single-layer graphene quantum dots with a similar size [20]. In the following we focus on this more symmetric regime, which moreover has been investigated in a dilution refrigerator with an electron temperature of around 100 mK. The increased energy resolution makes the edges of the triangles much more defined, such that the mutual capacitive coupling energy ($E_m$) between the two QDs can be extracted quantitatively. The coupling energy between both dots can be determined from the splitting of the triple points, $E_m = \alpha_{lg}^L \cdot \Delta V_{lg} - \alpha_{lg}^R \cdot \Delta V_{rg} = 2.4$ meV. (see Fig. 3b and arrows in Fig. 3c).

By changing the central gate voltage we are able to change the coupling energy between the left and right QD. Figures 4a and 4b show close-ups of the very same triple point ($V_b = 6$ mV) for two different inter-dot coupling energies $E_m = 4.8$ meV ($V_{cg} = 7.42$ V, Fig. 4a) and $E_m = 3.0$ meV ($V_{cg} = 7.54$ V, Fig. 4b), where the different triple point separations can be seen. Figure 4c presents a more detailed analysis of how the coupling energy changes with small variation in $V_{cg}$ for three individual nearby triple points (the examples shown in Figures 4a and 4b are highlighted by arrows). While we observe an increase of the coupling energy for the neighboring triple points [see (N,M) and (N+1,M), where N and M are the number of carriers on the left and right dot, respectively] for increasing $V_{cg}$ we observe a decrease of the coupling energy by more than a factor of 2 for the triple point (N+5,M-1). This is in agreement with earlier studies on graphene QDs [20, 22] showing a strongly non-monotonic dependence of the conductance on gate voltages due to the sharp resonances in the constrictions. Interestingly, the extracted coupling energies are roughly by a factor of 2 larger, as compared to earlier single-layer DQD studies [20, 22], which might be related to the con-
siderably smaller size of the investigated device.

The increased energy resolution also reveals additional fine structure inside the individual triple points. This, for example, can be seen in Figure 3c. The data exhibit distinct lines of increased conductance parallel to the base line of the triple points. Figure 4d shows a close-up of Figure 3c highlighting a set of electronic excited states. Inset: Schematic energy diagram showing the ground states of the double system and the first excited state of the left dot. (e,f) Different triple points featuring similar excited state spectra ($V_b = 10$ mV and 6 mV respectively). (g) Single-level spacing as a function of the relative change of the carrier occupation number on the left dot. The circles show the experimental data and the dashed horizontal line marks the experimental mean value ($\Delta = 1.82$ meV). The three curves represent the model for a single-layer QD for different absolute occupation numbers ($N = N_0 + \Delta N$) while the solid horizontal line marks constant level spacing for a bilayer dot. The QD diameter is assumed to be 50 nm.

In total we analyzed more than 50 excited states in a wide energy range. The electronic excited state energy spacing has been found to be constant over the entire range and a value of $\Delta = 1.75 \pm 0.27$ meV has been extracted. Interestingly this value is in good agreement with the single-particle confinement energy in disk-like bilayer graphene QDs, which can be estimated by using the density of states for bilayer graphene QDs, which can be estimated by using $\bar{E}(\bar{E}) = \gamma_1/F \sqrt{\bar{E}}$, where $v_F$ is the Fermi velocity and $\gamma_1 = 0.39$ meV is the inter-layer hopping energy. Consequently the single-particle level spacing is given by

$$\Delta = \frac{4\hbar^2 v_F^2}{\gamma_1} \frac{1}{d^2},$$

where $d$ is the diameter of the bilayer QD. Assuming a QD diameter of $d = 50$ nm, which is in reasonable agreement with the lithographically defined QD we obtain a constant (carrier number independent) level spacing of $\Delta = 1.71$ meV, which is in good agreement with the experimental data.

The charge carrier number independent level spacing is in contrast to single-layer graphene QDs where the single-particle level spacing, $\Delta(N) = \hbar v_F / d \sqrt{N}$, depends on the number of carriers, $N$. In Figure 4g we show the single-level spacing for a 50 nm diameter single-layer and
bilateral graphene QD as function of the excess number of carriers on the left dot, $\Delta N = N - N_0$. The solid horizontal line marks the constant level spacing for the bilayer QD, whereas the three $1/\sqrt{N}$-dependent curves belong to a single-layer QD with different absolute numbers of charge carriers on the dot ($N_0 = 5$, $25$ and $50$). The experimental data, which are depicted by circles in Fig. 4g have been extracted from charge stability diagrams as shown e.g. in Fig. 3c. For larger excess carrier numbers ($\Delta N > 40$) we also made use of relative gate lever arms to estimate $\Delta N$. Since we cannot fully exclude charge rearrangements, these data have to be taken with care. Please note that the total carrier numbers ($N, M$) on the two quantum dots are unknown. In Fig. 4g we plot only excited states energies extracted from measurements at $V_0 < 0$ and we focus on changes of the number of carriers on the left QD only ($\Delta N$, see also inset in Fig. 4d). For this limited set of excited states we find a constant value of $\Delta = 1.82 \pm 0.14$ meV, which is depicted by the dashed horizontal line in Fig. 4g. Please note that a constant level spacing might also result from an effective constant density of states induced by disorder. However, following Ref. 17, we estimate the effective dot disorder potential to be on the order of the addition energy of 5-10 charge carriers. Since we are probing a significantly larger energy range we can rule out disorder to be cause of the observed constant level spacing, and attribute it to true band structure properties. Consequently, the model suitable for describing single-layer QDs $[\Delta(N) \propto 1/\sqrt{N}]$ does not fit the experimental data for any reasonable $N_0$ whereas the model for a bilayer QD $[\Delta(N) = const.]$ is in good agreement. This leads to the conclusion that the investigated QDs are indeed extending over both graphene sheets forming the bilayer system.

Figure 5 shows a study of the evolution of excited states as function of the magnetic field oriented parallel to the bilayer graphene plane proving the electronic nature of these states. In Figures 5a and 5b we show a pair of triple points ($V_0 = -6$ mV) for two different values of magnetic field. Excited states parallel to the base line as well as one parallel to the left edge of the triangle are visible (dotted lines). For zero magnetic field again a level spacing of 1.8 meV (see e.g. arrow in Fig. 5a) can be observed. Figure 5b shows the very same triple points at a parallel magnetic field ($B_{\|}$) of 2 T. The position of the triangles in the gate voltage plane almost does not change at all (see crosses and solid lines in Fig. 5a,b). This holds even for $B_{\perp}$-fields up to 9 T (as shown in Fig. 5c) and shows that the effects on the orbital parts of the wave functions are unaffected. The parallel magnetic field has two effects on the triple points, i.e. triangles. First, the conductance at large detuning values slightly increases with increasing $B_{\perp}$-field. This is particularly true for the left triangle, where even parts of the triangle (left edge) are strongly suppressed at low $B_{\perp}$-fields. Since the associated slope differs from the actual triple point edge we attribute this effects to a modulated transmis-

![Figure 5](image-url) (color online) (a,b) The very same individual triple points recorded at $B_{\|} = 0$ T and $B_{\|} = 2$ T respectively ($V_0 = -6$ mV). (c) Current as a function of the detuning energy [measured along the dashed line indicated in panel (b)] for different $B_{\perp}$-fields up to 9 T. Traces are offset by 2 pA for clarity. (d) Current as a function of the detuning energy and the $B_{\|}$-field recorded at a different triple point. The corresponding triple point is shown in the inset, where the scale bars are 50 mV in gate voltages.

- sion from the drain to the right dot ($V_0 < 0$). Second, the 1st excited state parallel to the base line splits into two separated peaks (see dotted lines in Fig. 5b). This peak splitting is linear in magnetic field up to roughly 5 T as shown in Figure 5c, where we show current line cuts along the detuning energy axis (see dashed line in Fig. 5b). The different traces are recorded for different parallel magnetic fields (see labels at the right side) and they are offset by 2 pA in current for clarity. We observe that the position of the base line ($\varepsilon = 0$) and the 2nd excited state ($\varepsilon = 3.6$ meV) are constant up to 9 T as function of the $B_{\|}$-field (see vertical dashed lines in Fig. 5c). In contrast, the 1st excited state splits into two peaks, where the characteristic linear peak splitting is 0.2 meV/T (see diverging dashed lines). A similar peak splitting can also be observed on different triple points. In Figure 5d we show for example measurements taken on a triple point, where the 3rd excited state ($\varepsilon = 5.3$ meV) is splitting. Here the current at $V_0 = -8$ mV is plotted as a function of detuning energy and parallel magnetic field. The inset shows the corresponding triple points as well as the detuning axis (dashed line). While the position of the baseline and the 1st excited state are constant in $B_{\perp}$-field, the 2nd excited state shows a down-shift in energy and the 3rd excited state clearly splits linearly in
two peaks (see dashed lines). The base line as well as the 1st and 2nd excited state completely vanish at increased parallel magnetic fields. The observed B-field dependent peak shifts have again a slope of either 0 or ± 0.1 meV/T.

All observed peak splittings are likely to be originated from Zeeman spin splitting. This is motivated by the linear $B_{∥}$-field dependence of the peak splittings (up to high fields) and their characteristic energy scale, which is in reasonable agreement with $g_{\mu_B} = 0.116$ meV/T, assuming a $g$-factor of 2. The $g$-factor 2, moreover, would be in agreement with recent spin-resolved quantum interference measurements in graphene [35] and observations of spin states in a single-layer graphene quantum dot [19]. Moreover, we can not exclude that spin conserving inter-dot tunneling or different $g$-factors may be traced back to increased total spin numbers, non-known. Possible transitions leading to peak splittings may be traced back to increased total spin numbers, non-spin conserving inter-dot tunneling or different $g$-factors in both quantum dots. Moreover, we can not exclude that the local $g$-factor is increased by an unintentional layer-dependent doping or by the local gate potentials, which both could potentially lead to an out of plane electric field, which is expected to increase the effective $g$-factor in bilayer graphene [36].

In summary, we have fabricated and characterized a tunable bilayer graphene double quantum dot based on a width-modulated graphene nanostructure with lateral graphene gates. Its functionality was demonstrated by the observation of a tunable inter-dot coupling energy and the electronic excited state spectra over a wide energy range. We have shown that - in contrast to single-layer graphene - the single-particle level spacing is independent of the number of charge carriers on the bilayer quantum dots. By applying a $B_{∥}$-field we observed inter-dot transition energy splittings on the order of Zeeman splittings. These results give insights into tunable bilayer graphene double quantum dot devices and open the way to study individual spin states and spin coherence times in more detail in future experiments.

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