Spin qubits in graphene quantum dots

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The electron spin is a very promising candidate for a solid-state qubit [1]. Major experimental breakthroughs have been achieved in recent years using quantum dots formed in semiconductor heterostructures based on GaAs technology [2, 3, 4, 5]. In such devices, the major sources of spin decoherence have been identified as the spin-orbit interaction, coupling the spin to lattice vibrations [6, 7, 8], and the hyperfine interaction of the electron spin with the surrounding nuclear spins [9, 10, 11, 12, 13, 14]. Therefore, it is desirable to form qubits in quantum dots based on other materials, where spin-orbit coupling and hyperfine interaction are considerably weaker [15]. It is well known that carbon-based materials such as nanotubes or graphene are excellent candidates. This is so because spin-orbit coupling is weak in carbon due to its relatively low atomic weight, and because natural carbon consists predominantly of the zero-spin isotope $^{12}$C, for which the hyperfine interaction is absent. Here we show how to form spin qubits in graphene. A crucial requirement to achieve this goal is to find quantum dot states where the usual valley degeneracy is lifted. We show that this problem can be avoided in quantum dots with so-called armchair boundaries. We furthermore show that spin qubits in graphene can not only be coupled (via Heisenberg exchange) between nearest neighbor quantum dots but also over long distances. This remarkable feature is a direct consequence of the Klein paradox being a distinct property of the quasi-relativistic spectrum of graphene. Therefore, the proposed system is ideal for fault-tolerant quantum computation, and thus for scalability, since it offers a low error rate due to weak decoherence, in combination with a high error threshold due to the possibility of long-range coupling.

Only very recently, the fabrication of a single layer of graphene and the measurement of its electric transport properties have been achieved [16, 17, 18]. Two fundamental problems need to be overcome before graphene can be used to form spin qubits and to operate one or two of them as proposals in Refs. [1, 9]: (i) It is difficult to create a tunable quantum dot in graphene because of the absence of a gap in the spectrum. The phenomenon of Klein tunnelling makes it hard to confine quantum dots in graphene [19, 20, 21]. (ii) Due to the valley degeneracy that exists in graphene [22, 23, 24], it is non-trivial to form two-qubit gates using Heisenberg exchange coupling for spins in tunnel-coupled dots. Several attempts have been made to solve the problem (i) such as to use suitable transverse states in graphene ribbons to confine electrons [25], to combine single and bilayer regions of graphene [26], or to achieve confinement by using inhomogeneous magnetic fields [27]. The problem (ii) has not been recognized up to now. Here we propose a setup which solves both problems (i) and (ii) at once. Similar to Ref. [25] we choose to confine electrons by using suitable transverse states in a ribbon of graphene, cf. Fig. 1. In particular, we assume semiconducting armchair boundary conditions to exist on two opposite edges of the sample. It is by now feasible to experimentally identify ribbons of graphene with specific boundaries on the atomic scale. These are preferably of zigzag or of armchair type. For an experimental realization of our proposal, one would have to look for ribbons with semiconducting armchair boundaries. It is known that in such a device the valley degeneracy is lifted [28, 29], which is the essential prerequisite for the appearance of Heisenberg exchange coupling for spins in tunnel-coupled quantum dots (see below), and thus for the use of graphene dots for spin qubits. We show below that spin qubits in graphene can not only be coupled between nearest neighbor quantum dots but also over long distances. This long-distance coupling mechanism makes use of conduction band valence band tunnelling processes and is, therefore, directly based on the Klein paradox in graphene [20, 21].

We now discuss bound-state solutions in our setup, which are required for a localized qubit. We first concentrate on a single quantum dot which is assumed to be rectangular with width $W$ and length $L$, see Fig. 1. The basic idea of forming the dot is to take a ribbon of graphene with semiconducting armchair boundary conditions in $x$-direction and to electrically confine particles in $y$-direction. The low energy properties of electrons (with energy $\epsilon$ with respect to the Dirac point) in such a setup are described by the 4x4 Dirac equation

$$-i\hbar v \begin{pmatrix} \sigma_x \partial_x + \sigma_y \partial_y & 0 \\ 0 & -\sigma_x \partial_x + \sigma_y \partial_y \end{pmatrix} \Psi + eV(y)\Psi = e\Psi, \quad (1)$$

where the electric gate potential is assumed to vary stepwise, $V(y) = V_{\text{gate}}$ in the dot region (where $0 \leq y \leq L$), and $V(y) = V_{\text{barrier}}$ in the barrier region (where $y < 0$ or $y > L$). In Eq. (1), $\sigma_x$ and $\sigma_y$ are Pauli matrices, $\hbar$ is Planck’s constant divided by $2\pi$, $v$ the Fermi velocity, and $e$ the charge of an electron. The four component spinor envelope wave function $\Psi = (\Psi_A^{(K)}, \Psi_B^{(K)}, -\Psi_A^{(K)}, -\Psi_B^{(K)})$ varies on scales large compared to the lattice spacing. At this point, we are only interested in the orbital structure of the wave function. The spin degree of freedom is neglected until the final part, where we discuss the Heisenberg exchange coupling for spins in tunnel-coupled quantum dots. In the wave function $\Psi, A$ and $B$ refer to the two sublattices in the two-dimensional honeycomb lattice of carbon atoms, whereas $K$ and $K'$ refer to the vectors $\mathbf{K}$ and $\mathbf{K}'$ in reciprocal space corresponding to the two valleys in the bandstructure of graphene. The appropriate semiconducting armchair boundary conditions for such a wave function have been formulated in Ref. [28] and can be written as ($\alpha = A, B$)

$$\Psi^{(K')}_{\alpha}|_{x=0} = \Psi^{(K')}_{\alpha}|_{x=W}, \quad \Psi^{(K')}_{\alpha}|_{x=L} = e^{i2\pi/3}\Psi^{(K')}_{\alpha}|_{x=0}, \quad \Psi^{(K')}_{\alpha}|_{x=0} = e^{i2\pi/3}\Psi^{(K')}_{\alpha}|_{x=W}, \quad (2)$$

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corresponding to a width $W$ of the ribbon shown in Fig. 1 where $W$ is not an integer multiple of three unit cells. The ± signs in Eq. (4) (as well as in Eq. (3) below) correspond to the two possible choices of a number of unit cells that is not an integer multiple of three. The full set of plane wave solutions of Eq. (1) is readily determined [29]. It is well known that the boundary condition (2) yields the following quantization conditions for the wave vector $k_y \equiv q_y$ in $x$-direction [28, 29]

\[ q_n = (n \pm 1/3)\pi/W, \quad n \in \mathbb{Z}. \]  

(3)

An explicit form of the corresponding wave functions is presented in App. A and App. B. The level spacing of the modes [3] can be estimated as $\Delta \varepsilon \approx \hbar v/3W$, which gives $\Delta \varepsilon \approx 30$ meV, where we used that $v \approx 10^6$ m/s and assumed a quantum dot width of about $W \approx 30$ nm. Note that Eq. (3) also determines the energy gap for excitations as $E_{\text{gap}} = 2\hbar v q_0$. Therefore, this gap is of the order of 60 meV, which is unusually small for semiconductors. This is a unique feature of graphene that will allow for long-distance coupling of spin qubits as will be discussed below.

We now present in more detail the ground-state solutions, i.e. $n = 0$ in Eq. (3). The corresponding ground-state energy $\varepsilon$ can be expressed relative to the potential barrier $V = V_{\text{barrier}}$ in the regions $y < 0$ and $y > L$ as $\varepsilon = \varepsilon_{\text{barrier}} \pm \hbar v (q_y^2 + k^2)^{1/2}$. Here, the ± sign refers to a conduction band (+) and a valence band (−) solution to Eq. (1). For bound states to exist and to decay at $y \to \pm \infty$, we require that $\hbar v q_0 > |\varepsilon - \varepsilon_{\text{barrier}}|$, which implies that the wave vector $k_y \equiv k$ in $y$-direction, given by

\[ k = i \sqrt{q_y^2 - (|\varepsilon - \varepsilon_{\text{barrier}}|/\hbar)^2}, \]  

(4)

is purely imaginary. In the dot region (0 $\leq y \leq L$), the wave vector $k$ in $y$-direction is replaced by $\tilde{k}$, satisfying $\varepsilon = eV_{\text{gate}} \pm \hbar v (q_y^2 + \tilde{k}^2)^{1/2}$. Again the ± sign refers to conduction and valence band solutions. In the following, we focus on conduction band solutions to the problem.

Since the Dirac equation (1) implies the continuity of the wave function, the matching condition at $y = 0$ and $y = L$ allows us to derive the transcendental equation for $\varepsilon$

\[ e^{2\tilde{k}L}(z_{0,\tilde{k}} - z_{0,k})^2 - (1 - z_{0,k}z_{0,\tilde{k}}) = 0 \]  

(5)

with $z_{0,\tilde{k}} \equiv (q_0 + i\tilde{k})/(q_0^2 + \tilde{k}^2)^{1/2}$. Eq. (5) determines the allowed energies $\varepsilon$ for bound states. In order to analyze the solutions to Eq. (5), we distinguish two cases, one where $\tilde{k}$ is real, and the other, where $\tilde{k}$ is purely imaginary. The two cases are distinguished by the condition $|\varepsilon - eV_{\text{gate}}| \geq \hbar v q_0$ and $|\varepsilon - eV_{\text{gate}}| < \hbar v q_0$, respectively. Furthermore, we assume that $V_{\text{gate}} \neq V_{\text{barrier}}$, i.e., $z_{0,\tilde{k}} \neq z_{0,k}$. If we relax this assumption, we can show that for the case $z_{0,k} = z_{0,\tilde{k}}$ only a single solution to Eq. (5) exists, namely $z_{0,\tilde{k}} = 1$, which implies that $\tilde{k} = 0$. The corresponding wave function to this solution vanishes identically (see App. A for further details). In the case where $\tilde{k}$ is purely imaginary, there is no bound-state solution. This is due to the fact that such a solution would have to exist directly in the bandgap. We now analyze solutions for real $\tilde{k}$.

In the corresponding energy window

\[ |\varepsilon - eV_{\text{gate}}| \geq \hbar v q_0 > |\varepsilon - eV_{\text{barrier}}|, \]  

(6)

we can simplify Eq. (5) considerably, obtaining

\[ \tan(\tilde{k}L) = \frac{\hbar \sqrt{2(\hbar v q_0)^2 - (\varepsilon - eV_{\text{barrier}})^2}}{(\varepsilon - eV_{\text{barrier}})(\varepsilon - eV_{\text{gate}}) - (\hbar v q_0)^2}. \]  

(7)

We show a set of solutions to Eq. (7) for a relatively short dot ($q_0L = 2$) as well as a longer dot ($q_0L = 5$) in Fig. 2. The number of bound states $N$ (for $n = 0$) is maximal if $\Delta V = V_{\text{barrier}} - V_{\text{gate}}$ is exactly as large as the size of the gap $E_{\text{gap}} = 2\hbar v q_0$, then $N_{\text{max}} = \left\lceil \sqrt{8q_0L/\pi} \right\rceil$, where $[x]$ is the integer just larger than $x$. The level spacing associated with the allowed solutions of Eq. (7) increases as $L$ decreases and has a rather complicated parameter dependence. It can, however, be estimated to be of the order of $\Delta \varepsilon \approx \hbar v / \pi L$, which is in the energy range of a few tens of meV as mentioned below Eq. (3). In Fig. 3 we show the energy bands of a single dot and two neighboring barrier regions as well as a double dot setup with three barrier regions. The double dot case illustrates how we make use of the Klein paradox to couple two dots.

A particular example of a wave function is shown in Fig. 4. It is a ground-state solution under the parameter choice $e(V_{\text{barrier}} - V_{\text{gate}}) = 0.6\hbar v q_0$, and $q_0L = 2$ (indicated by the arrow in Fig. 2). The weight of the wave function on the $A$ and $B$ lattice sites is different, however, the integrated weight is the same as required by the normalization condition [28].

Ground-state solutions (i.e. the lowest lying (red) lines in Fig. 3) have no nodes in the dot region – similar to the corresponding problem of confined electrons that obey the non-relativistic Schrödinger equation. Excited-state solutions in
FIG. 2: Bound state solutions for two different dot sizes. Bound-state solutions of a relatively long ($q_0L = 5$, left panel) and a shorter ($q_0L = 2$, right panel) quantum dot are shown. The diagonal straight lines mark the area in which bound-state solutions can occur. The arrow marks the solution for which the wave function is plotted in Fig. 4.

parameter regions in which they exist, do have nodes in the dot region, which is shown in Fig. 8 of App. C.

We now turn to the case of two coupled graphene quantum dots, separated by a potential barrier, as sketched in Fig. 1 each dot filled with a single electron. It is interesting to ask whether the spins $S_i$ of these two electrons ($i = 1, 2$) are coupled through an exchange coupling, $J_{\text{exch}} = J S_i \cdot S_j$, in the same way as for regular semiconductor quantum dots [9], because this coupling is, in combination with single-spin rotations, sufficient to generate all quantum gates required for universal quantum computation [1]. The exchange coupling is based on the Pauli exclusion principle which allows for electron hopping between the dots in the spin singlet state (with opposite spins) of two electrons, but not in a spin triplet (with parallel spins), thus leading to a singlet-triplet splitting (exchange energy) $J$. However, a singlet-triplet splitting $J \neq 0$ only occurs if the triplet state with two electrons on the same dot in the ground state is forbidden, i.e., in the case of a single non-degenerate orbital level. This is a non-trivial requirement in a graphene structure, as in bulk graphene, there is a two-fold orbital (“valley”) degeneracy of states around the points $K$ and $K'$ in the first Brillouin zone. This valley degeneracy is lifted in our case of a ribbon with semiconducting armchair edges, and the ground-state solutions determined by Eq. (7) are in fact non-degenerate. The magnitude of the exchange coupling within a Hund-Mulliken model is $J = (-U_t + (U_t^2 + 16U_H^2)^{1/2})/2 + V$, where $t$ is the tunnelling (hopping) matrix element between the left and right dot, $U$ is the on-site Coulomb energy, and $V$ is the direct exchange from the long-range (inter-dot) Coulomb interaction. The symbols $U_t$ and $U_H$ indicate that these quantities are renormalized from the bare values $t$ and $U$ by the inter-dot Coulomb interaction. For $t \ll U$ and neglecting the long-ranged Coulomb part, this simplifies to the Hubbard model result $J = 4t^2/U$ where $t$ is the tunnelling (hopping) matrix element between the left and right dot and $U$ is the on-site Coulomb energy. In the regime of weak tunnelling, we can estimate $t \approx \varepsilon \int W_L(x,y)\Psi_R(x,y)dx\,dy$, where $\Psi_L(x,y) = \Psi(x,y \pm (d + L)/2)$ are the ground-state spinor wave functions of the left and right dots and $\varepsilon$ is the single-particle ground state energy. Note that the overlap integral vanishes if the states on the left and right dot belong to different transverse quantum numbers $q_{n_{\perp}} \neq q_{n_{\perp}'}$. For the ground state mode, we have $n_{\perp} = n_{\perp}' = 0$, and the hopping matrix element can be estimated for $d \gtrsim L$ as

$$t \approx 4\varepsilon \alpha_0 \delta_0 W_{x_{0,L}} \exp(-d/k),$$

where $\alpha_0$ and $\delta_0$ are wave function amplitudes (with dimension $1/\text{length}$) that are specified in App. C. As expected, the exchange coupling decreases exponentially with the barrier thickness, the exponent given by the “forbidden” momentum $k$ in the barrier, defined in Eq. (4). The amplitude $t$ can be maximized by tuning to a bound-state solution, where
FIG. 4: **Ground-state wave function.** Normalized squared wave functions $|\Psi_A|^2 = |\Psi_{A_1}|^2 = |\Psi_{A_2}|^2$ and $|\Psi_B|^2 = |\Psi_{B_1}|^2 = |\Psi_{B_2}|^2$ for the bound state solution for the parameter choice $e(V_{\text{barrier}} - V_{\text{gate}}) = 0.6\hbar v q_0$, and $q_0 L = (\pi/3) L/W = 2$ (indicated by the arrow in Fig. 2). The corresponding energy is given by $\varepsilon \approx 1.31 \cdot \hbar v q_0$. The dotted lines indicate the dot region $0 \leq y \leq L$.

$|\varepsilon - eV_{\text{barrier}}|$ approaches $\hbar v q_0$ (from below). Then, $d|k| < 1$. Such a fine-tuning can be easily achieved in graphene quantum dots, where the small band gap allows to sweep through it, and, therefore, to use conduction and valence band states (of the barrier region) to couple quantum dots. In Fig. 3(b), we sketch the energy bands for the double dot case which shows how confined states in the two dots can be coupled via cotunnelling processes through the continuum of states in the valence band of the central barrier region. Remarkably, this opens up the possibility for long distance coupling of electron spins because, in the limit $|\varepsilon - eV_{\text{barrier}}| \rightarrow \hbar v q_0$, the coupling $t$ depends only weakly on the distance $d$ between the quantum dots. However, already for bound state solutions with $|k|d > 1$, a coupling over a length exceeding several times the dot size is possible. For the situation where we couple two ground states in the quantum dots, we find, for instance, a solution, where $|k|d = 4$, $d = 10 L$, and the coupling can still be as large as $t \approx 0.03 e$, for highly localized qubits. This example is shown in Fig. 5. If we couple a ground state in the one dot with an excited state in the other dot, the hopping matrix element $t$ can be even larger. The corresponding functions for that case are illustrated in Fig. 5. The values of $t$, $U$, and $J$ can be estimated as follows. The tunnelling matrix element $t$ is a fraction of $\varepsilon \approx 30 \text{ meV}$ (for a width of $W \approx 30 \text{ nm}$), we obtain that $t \approx 0.5 \ldots 2.5 \text{ meV}$. The value for $U$ depends on screening which we can assume to be relatively weak in graphene [24], thus, we estimate, e.g., $U \approx 10 \text{ meV}$, and obtain $J \approx 0.1 \ldots 1.5 \text{ meV}$.

For the situation with more than two dots in a line, it turns out that we can couple any two of them with the others being decoupled by detuning. We mention here that our model is based on a single particle picture. Such a model effectively captures effects of Coulomb interactions as far as they can be described within the constant interaction model for quantum dots [30]. The Coulomb interaction then only shifts the energy levels in each dot by a constant. In Fig. 6, we illustrate the situation of three dots in a line where the left and the right dot are strongly coupled and the center dot is decoupled by detuning. The tunnel coupling of dot 1 and dot 3 is then achieved via Klein tunneling through the valence band of the two central barriers and the valence band of the center dot. It is important for the long-distance coupling that the exchange coupling of qubit 1 and qubit 3 is primarily achieved via the valence band and not via the qubit level of the center dot – leaving the qubit state of dot 2 unchanged. Using the standard transition matrix approach, we can compare the transition rate of coupling dot 1 and dot 3 via the continuum of states in the valence band of...
the center dot (which we call \( \Gamma_{\text{VB}} \)) with the transition rate via
the detuned qubit level of the center dot (which we call \( \Gamma_{\text{QB}} \)).
We obtain for the ratio (see App. D for the derivation)
\[
\frac{\Gamma_{\text{VB}}}{\Gamma_{\text{QB}}} \approx \left( \frac{L}{W} \right) \ln(4L/E_{\text{gap}}),
\]
where \( \Delta \approx 6 \text{ eV} \) is the band width of graphene. Therefore,
by increasing the aspect ratio \( L/W \), it is possible to in-
crease the rate \( \Gamma_{\text{VB}} \) with respect to \( \Gamma_{\text{QB}} \). For \( L/W = 2 \)
and \( E_{\text{gap}} = 60 \text{ meV} \), we find that \( \Gamma_{\text{VB}}/\Gamma_{\text{QB}} \approx 12 \), mean-
ing that the qubit level in dot 2 is barely used to couple dot 1 and dot
3. This is a unique feature of graphene quantum dots due to
the small and highly symmetric band gap, which is not known
to exist for other semiconducting materials. The availability of
non-local interactions is important in the context of quantum
error correction, as it raises the error threshold for fault-
tolerant quantum computation \[31\]. In conclusion, we have
proposed a setup to form spin qubits in quantum dots based on
graphene nanoribbons with semiconducting armchair bound-
daries. For such a system, we have calculated bound states of a
tunable dot and outlined how two-qubit gates can be realized.
We expect very long coherence times for such spin qubits
since spin-orbit coupling and hyperfine interaction are known
to be weak in carbon, see App. E. Furthermore, we have found
that the high flexibility in tuning graphene quantum dots in
combination with conduction band to valence band tunnelling
based on the Klein paradox allows for long distance coupling
electron spins. Therefore, we propose a system which can
serve as the fundamental building block for scalable and fault-
tolerant quantum computing.

APPENDIX A: GENERAL MODEL

We now present in detail how to derive solutions for bound
states in a graphene quantum dot. The dot is assumed to be
rectangular with width \( W \) and length \( L \) as illustrated in
Fig. 1. The basic idea of forming the dot is to take a strip of
graphene with semiconducting armchair boundary condi-
tions in \( x \)-direction and to electrically confine particles in \( y \)-
direction. Transport properties of a similar system have been
discussed in Ref. \[25\].

The low energy properties of electrons with energy \( \epsilon \)
in such a setup are described by the 4x4 Dirac equation
\[
-\text{i} \hbar v \begin{pmatrix}
\sigma_x \partial_x + \sigma_y \partial_y & 0 \\
0 & -\sigma_x \partial_x + \sigma_y \partial_y
\end{pmatrix} \Psi + eV(y)\Psi = e\Psi,
\]
with the electric gate potential
\[
V(y) = \begin{cases}
V_{\text{gate}}, & (0 \leq y \leq L), \\
V_{\text{barrier}}, & \text{otherwise}.
\end{cases}
\]
In Eq. (A1), \( \sigma_x \) and \( \sigma_y \) are Pauli matrices, \( \hbar \) is Planck’s con-
stant divided by 2\( \pi \), \( e \) is the electron charge, and \( v \) is the
Fermi velocity. The four component envelope wave function
\( \Psi = (\Psi_A^{(K)}, \Psi_B^{(K)}, -\Psi_A^{(K')}, -\Psi_B^{(K')}) \) varies on scales large
compared to the lattice spacing. Here, \( A \) and \( B \) refer to the two
sublattices in the two-dimensional honeycomb lattice of car-
bon atoms, whereas \( K \) and \( K' \) refer to the vectors in reciprocal
space corresponding to the two valleys in the bandstructure of
graphene.

Plane wave solutions to Eq. (A1) take the form \[29\]
\[
\Psi_{n,k}^{(x,y)}(x,y) = \chi_{n,k}^{(x)}(x)e^{iyk}, \quad \Psi_{n,k}^{(-x,y)}(x,y) = \chi_{n,k}^{(-x)}(x)e^{-iyk}
\]
with
\[
\chi_{n,k}^{(x)}(x) = a_{n,+} e^{iq_n x} + a_{n,-} e^{-iq_n x},
\]
\[
\chi_{n,k}^{(-x)}(x) = a_{n,-} e^{iq_n x} + a_{n,+} e^{-iq_n x},
\]
and
\[
\chi_{n,k}^{(y)}(y) = a_{n,+} e^{iq_n y} + a_{n,-} e^{-iq_n y},
\]
\[
\chi_{n,k}^{(-y)}(y) = a_{n,-} e^{iq_n y} + a_{n,+} e^{-iq_n y}.
\]
The complex number \( z_{n,k} \) is given by
\[
z_{n,k} = \pm \frac{q_B + ik}{\sqrt{q_B^2 + k^2}}.
\]
The energy of the state in the barrier regions \( y < 0 \) and \( y > L, \)
where \( V = V_{\text{barrier}} \) is given by
\[
\epsilon = eV_{\text{barrier}} + \hbar v \sqrt{q_B^2 + k^2}.
\]
In the dot \( 0 \leq y \leq L \), where \( V = V_{\text{gate}} \) the wave vector \( k \) is
replaced by \( \tilde{k} \), satisfying
\[
\epsilon = eV_{\text{gate}} + \hbar v \sqrt{q_B^2 + \tilde{k}^2}.
\]
The \( \pm \) sign in Eqs. (A6) – (A8) refers to conduction and va-
lence bands. In the following, we concentrate on conduction
band solutions of the problem (keeping in mind that there is
always a particle-hole conjugated partner solution).
The transverse wave vector \( q_B \) as well as the coefficients
\( a_{n,+}, a_{n,-}, b_{n,+}, b_{n,-} \) of the \( n \)-th mode are determined (up to a
normalization constant) by the boundary conditions at \( x = 0 \)
and \( x = W \). We consider a class of boundary conditions for
which the resulting parameters are independent of the longi-
tudinal wave vectors \( k \) and \( \tilde{k} \). We are particularly interested
in semiconducting armchair boundary conditions defined by
\[28\]
\[
\Psi|_{x=0} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \Psi|_{x=0},
\]
\[
\Psi|_{x=W} = \begin{pmatrix} 0 & e^{-2\pi i/3} \\ e^{2\pi i/3} & 0 \end{pmatrix} \Psi|_{x=W}.
\]
where $\mu = \pm 1$ is defined by the width of the graphene strip $W = a_0 (3M + \mu)$, with $M$ a positive integer ($a_0 = 0.246$ nm is the graphene lattice constant), and $\textbf{I}$ is the 2x2 unit matrix. A strip whose width is an integer multiple of three unit cells ($\mu = 0$) is metallic and not suitable for spin qubit applications. The states of a semiconductor strip are non-degenerate (in valley space):

\[ q_n = \frac{\pi}{W} (n + \mu/3), \quad n \in \mathbb{Z} \]  

(A11)

with $a_{n,k} = b'_{n,k} = 0$, $a_{n,k} = b_{n,k}$ (for $\mu = -1$) or $a'_{n,k} = b_{n,k} = 0$, $a_{n,k} = b'_{n,k}$ (for $\mu = 1$). Note that $q_n$ determines the size of the gap for each mode $n$ that is due to the boundary conditions. The size of the gap of mode $n$ is given by $2 \hbar q_n$. For concreteness, we consider the case of $\mu = 1$ only. It can be shown that for the case of $\mu = -1$, the bound states and normalized squared wave functions have exactly the same dependence on the parameters of a quantum dot, $V_{\text{barrier}}$, $V_{\text{gate}}$, $W$, and $L$.

Our ansatz for a bound state solution at energy $\varepsilon$ to Eq. (1) then reads

\[ \Psi = \begin{cases} \alpha_n \gamma_n(x) e^{-iky}, & \text{if } y < 0, \\ \beta_n \gamma_n(x) e^{iky} + \gamma_n \alpha_n \gamma_n(x) e^{-iky}, & \text{if } 0 \leq y \leq L, \\ \delta_n \gamma_n(x) e^{iky - L}, & \text{if } y > L. \end{cases} \]  

(A12)

For bound states, the wave function should decay for $y \to \pm \infty$, so we require that

\[ k = i \sqrt{2q_n^2 - (\varepsilon - eV_{\text{barrier}})^2/(\hbar^2)}, \]  

(A13)

where $\hbar q_n > |\varepsilon - eV_{\text{barrier}}|$ always has to hold.

To find bound state solutions, we have to analyze the following set of equations (coming from wave function matching at $y = 0$ and $y = L$)

\[ \begin{align*} 
\alpha_n \left( z_{n,k} \right) & = \beta_n \left( z_{n,k} \right) + \gamma_n \left( \tau_{n,k} \right), \\
\delta_n \left( z_{n,k} \right) & = \beta_n \left( 1 \right) e^S + \gamma_n \left( \tau_{n,k} \right) e^{-S},
\end{align*} \]  

(A14)

where $\alpha_n = \alpha_n a_{n,-}, \beta_n = \beta_n a_{n,+}, \gamma_n = \gamma_n a_{n,-}, \delta_n = \delta_n a_{n,+}$, and $S \equiv ikL$. We can write Eq. (A14) as

\[ \begin{pmatrix} z_{n,k} & -1 & -z_{n,k} & 0 \\ 1 & -z_{n,k} & 1 & 0 \\ 0 & -e^S & -z_{n,k} & 1 \\ 0 & -z_{n,k} e^S & 1 & 0 \end{pmatrix} \begin{pmatrix} \alpha_n \\ \beta_n \\ \gamma_n \\ \delta_n \end{pmatrix} = 0. \]  

(A15)

The allowed energy values $\varepsilon$ are readily determined by finding the roots of the determinant of the matrix on the lhs of the latter equation

\[ e^{2S} (z_{n,k} - z_{n,k})^2 - (1 - z_{n,k} z_{n,k})^2 = 0. \]  

(A16)

A rather obvious solution is $z_{n,k} = 1$ (which implies that $S = 0$) corresponding to

\[ \varepsilon = \pm \hbar q_n + eV_{\text{gate}}. \]  

(A17)

However, the corresponding wave functions to the allowed energy solutions (A17) vanish identically. So, in order to proceed, we have to find other (less trivial) solutions to the transcendental equation (A16). Since the case $z_{n,k} = z_{n,k}$ only has the trivial solution (A17), we can assume that $z_{n,k} \neq z_{n,k}$, which means that $V_{\text{barrier}} \neq V_{\text{gate}}$. Then, we find that

\[ e^S = \pm \frac{1}{z_{n,k} - z_{n,k}}. \]  

(A18)

To analyze the solutions of Eq. (A18), we distinguish two cases, one where

\[ S = ikL \]  

(A19)

is purely imaginary (i.e., $\bar{k}$ is real), and another, where $S$ is real. The two cases are distinguished by the criterion $|\varepsilon - eV_{\text{gate}}| \geq \hbar q_n$ and $|\varepsilon - eV_{\text{barrier}}| < \hbar q_n$, respectively. In the former case, since the lhs of Eq. (A18) has modulus unity, also the rhs must be unimodular, which is satisfied if in addition $|\varepsilon - eV_{\text{barrier}}| \leq \hbar q_n$. This case, where the equation for the argument of (A18) remains to be solved, is discussed in Sec. B. The latter case where $S$ is real has no solutions.

Indeed, let us rewrite Eq. (A18) as follows:

\[ e^{2S} - 1 = -\frac{2\bar{k}k}{q_n^2 - (\varepsilon - eV_{\text{barrier}})(\varepsilon - eV_{\text{gate}})/\hbar^2} + \bar{k}k \]  

(A20)

Taking into account that $q_n^2 > (\varepsilon - eV_{\text{barrier}})(\varepsilon - eV_{\text{gate}})/\hbar^2$ and $\bar{k}k \in \mathbb{R}$, we find that the left and right sides of this equation have different signs, therefore, Eq. (A18) has no roots for any purely imaginary $\bar{k}$.

**APPENDIX B: BOUND STATE SOLUTIONS**

We now restrict ourselves to the energy window

\[ |\varepsilon - eV_{\text{gate}}| \geq \hbar q_n \geq |\varepsilon - eV_{\text{barrier}}|. \]  

(B1)

Then $\bar{k}$ is real, therefore $|\varepsilon^2| = 1$ and $|z_{n,k}| = 1$. Furthermore, $z_{n,k}$ is real. We define $z_{n,k} \equiv e^{i\theta_n}$, where

\[ \theta_n = \arctan(\bar{k}/q_n). \]  

(B2)

It is easy to verify that in the energy window (B1)

\[ \frac{1 - z_{n,k} \tau_{n,k}}{z_{n,k} - \bar{z}_{n,k}} = 1. \]  

(B3)

We can now rewrite Eq. (A18) as

\[ \tan(\bar{k}L) = \frac{\sin \theta_n (1 - z_{n,k}^2)}{2z_{n,k} \tau_{n,k} - 1 + z_{n,k}^2 \cos \theta_n} \]  

(B4)

and further simplify this expression by using that

\[ \sin \theta_n = \frac{\bar{k}/q_n}{\sqrt{1 + (\bar{k}/q_n)^2}}, \]  

(B5)

\[ \cos \theta_n = \frac{1}{\sqrt{1 + (\bar{k}/q_n)^2}}. \]  

(B6)
After some algebra, we obtain

$$\tan(kL) = \frac{-ik}{(\epsilon - eV_{\text{barrier}})(\epsilon - eV_{\text{gate}})/(\hbar v)^2 - q_n^2}. \quad (B7)$$

The latter equation in combination with Eq. (A13) yields Eq. (7). Numerical solutions to Eq. (B7) are shown in Fig. 2.

By applying different voltages to the gate and the barriers we shift the energy bands of the graphene ribbon under the barriers with respect to that of the quantum dot. A bound state in the quantum dot is allowed once the energy of the state hits the band gap of the barriers. If the difference of the barrier and gate voltages $\Delta V = |V_{\text{barrier}} - V_{\text{gate}}|$ is less than the energy of the gap $2\hbar v q_n$ for $n$-th subband, $k$ of a bound state lies in the interval $[-k_{\text{max}}, k_{\text{max}}]$, where $k_{\text{max}}$ is found from the condition $\epsilon(k_{\text{max}}) = V_{\text{barrier}} + \hbar v q_n$ and, therefore, $k_{\text{max}} = e\Delta V \sqrt{1 + 2\hbar v q_n/k \Delta V}$. The number of bound states (for a given subband index $n$) is proportional to the length of the quantum dot $L$ and is given by $N = \left[ k_{\text{max}}/L/\pi \right] (|x|) = \int n \pi/|x|$. The number of the bound states of the $n$-th subband is maximal, when the barrier-gate voltage difference equals the energy band gap ($\Delta V = 2\hbar v q_n$), and so $N_{\text{max}} = \left[ \sqrt{8} q_n L/\pi \right]$. In the case of $\Delta V > 2\hbar v q_n$, the top of the valence band of the graphene ribbon under the barriers becomes higher than the bottom of the conduction band of the quantum dot, therefore, there are no bound states with energies $\epsilon < eV_{\text{barrier}} - \hbar v q_n$ and $|k|$ of a bound state lies in the interval $[-k_{\text{min}}, k_{\text{max}}]$, where $k_{\text{min}}$ is found from the condition $\epsilon(k_{\text{min}}) = V_{\text{barrier}} - \hbar v q_n$ ($\kappa_{\text{min}} = e\Delta V \sqrt{1 - 2\hbar v q_n/\Delta V}$), therefore, bound states lie in the energy window $eV_{\text{barrier}} - \hbar v q_n \leq \epsilon \leq eV_{\text{barrier}} + \hbar v q_n$ (as shown in Fig. 2) and the number of the bound states is given by $N = \left[ k_{\text{max}}/L/\pi \right] - \left[ k_{\text{min}}/L/\pi \right]$.

With increasing the barrier-gate voltage difference, a $m$-th bound state appears at $\Delta V_0 = -\hbar v q_n + \hbar \sqrt{q_n^2 + (\pi L)^2 (m - 1)^2}$, with the energy $\epsilon_m = V_{\text{barrier}} + \Delta V_0 + \hbar v q_n$ and ends up at $\Delta V_1 = \hbar v q_n + \hbar \sqrt{q_n^2 + (\pi L)^2 m^2}$ with the energy $\epsilon_m = V_{\text{gate}} + \Delta V_1 - \hbar v q_n$ (see Fig. 2).

**APPENDIX C: WAVE FUNCTION**

Following Brey and Fertig [28], we write the wave function as

$$\Psi(x, y) = \begin{pmatrix} \Psi_A^K(x, y) \\ \Psi_B^K(x, y) \\ -\Psi_A^{K'}(x, y) \\ -\Psi_B^{K'}(x, y) \end{pmatrix} \quad (C1)$$

and give solutions for each component separately. As mentioned above, the subscripts $A$ and $B$ refer to the two sublattices in the two-dimensional honeycomb lattice of carbon atoms and the superscripts $K$ and $K'$ refer to the two valleys in graphene. Note that the normalization condition [28]

$$\int dxdy \left[ |\Psi_{\mu}^{(K)}(x, y)|^2 + |\Psi_{\mu}^{(K')}(x, y)|^2 \right] = \frac{1}{2} \quad (C2)$$

In the latter equations, the first line corresponds to the region in space, where $y < 0$, the second line to $0 \leq y \leq L$, and the third line to $y > L$. Thus, we obtain that

$$|\Psi_{\mu}^{(K)}(x, y)|^2 = |\Psi_{\mu}^{(K')}(x, y)|^2, \quad (C7)$$

$$|\Psi_{\mu}^{(K)}(x, y)|^2 = |\Psi_{\mu}^{(K)}(x, y)|^2. \quad (C8)$$

We now plot the normalized squared wave function of a ground-state solution and an excited-state solution of a dot with length $q_n L = 5$ in Figs. 7 and 8 respectively. These are obtained from Fig. 2 under the choice that $e(V_{\text{barrier}} - V_{\text{gate}}) = 0.5 \hbar v q_n$. Evidently, the ground-state solution has no nodes in the dot region, whereas the excited-state solution has nodes.

![FIG. 7: Ground-state wave function. Normalized squared wave function $|\Psi|^2 = |\Psi_A^K|^2 = |\Psi_B^K|^2$ and $|\Psi_B|^2 = |\Psi_A^{K'}|^2$ for the ground state solution of a dot of length $q_n L = 5$ with corresponding energy $\epsilon = 1.101 \hbar v q_n$. Here, $e(V_{\text{barrier}} - V_{\text{gate}}) = 0.5 \hbar v q_n$. The dotted lines indicate the dot region $0 \leq y q_n \leq 5$.](image-url)
APPENDIX D: LONG-DISTANCE COUPLING

1. Long-distance coupling of two qubits

Here, we discuss a particular example of long-distance coupling of two qubits separated by a distance $d$. The coupling is achieved via a continuum of states in the valence band of the barrier region as shown in Fig.3(b) of the Letter. Therefore, the long-range coupling is enabled by the Klein paradox. In the weak tunneling regime, the hopping matrix element is given by

$$ t \approx \epsilon \int \Psi_L^\dagger(x,y)\Psi_R(x,y)dx\,dy, $$

where $\Psi_{L,R}(x,y) = \Psi(x,y \pm (d + L)/2)$ are the spinor wave functions of the left and right dots and $\epsilon$ is the single-particle energy of the coupled levels. The integration in transverse $x$-direction is trivial and just gives a factor $W$. The integration in longitudinal $y$-direction can be restricted to the integration window $y \in [-d/2, d/2]$ if the wave functions are predominantly localized in the dot regions. Then, the hopping matrix element can be estimated for $d \gtrsim L$ as

$$ t \approx 4\epsilon d_0^2 Wd_{\alpha}\exp(-d|k|), $$

where $\alpha_0$ and $d_0$ are wave function amplitudes specified in Eqs. (C3) – (C6). In Eq. (D2), we assumed that only levels of the series corresponding to the $n = 0$ transverse mode are coupled. It is easy to relax this assumption because, if higher transverse modes form bound states, then only modes with $n_L = n_R$ contribute to $t$, where $n_{L,R}$ is the transverse quantum number in the left/right dot. In Fig. 9, we demonstrate that a rather large coupling of $t = 0.06\epsilon$ can be achieved over a distance as large as ten times the size of the quantum dots (see also Fig. 5 for comparison). Note that the qubits in this example are well localized in the corresponding dot regions: The probability of the electron in the left dot to be in the dot region $P(\text{in})$ is 7.2 times larger than to be in the barrier regions $P(\text{out})$. For the right dot, the ratio of $P(\text{in})/P(\text{out}) = 2.7$ is a bit smaller but the electron is still predominantly localized in the dot region.

2. Long-distance coupling in multiple quantum dot setup

In Fig.6, we propose a triple quantum dot setup in which dot 1 and dot 3 are strongly coupled and the center dot 2 is decoupled by detuning. It is important that dot 1 and dot 3 are coupled via the valence band states of dot 2 and not via the (detuned) qubit level of dot 2. Otherwise, the spin of the decoupled qubit level would be affected by the coupling of the other qubits which is unwanted in the proposed long-distance coupling scheme. We assume that the gates that put the three dots in the Coulomb blockade regime are set in such a way that cotunnelling processes from dot 1 via dot 2 to dot 3 happen in the following order: First, an electron tunnels from dot 2 to dot 3 and then an electron tunnels from dot 1 to dot 2. The system is described by the Hamiltonian

$$ H = H_0 + H_T, $$

where the kinetic term describes three qubit levels ($\alpha = 1, 2, 3$) and the continuum of states in the valence band of dot 2

$$ H_0 = \sum_{\alpha=1-3} \sum_{\sigma = \uparrow, \downarrow} E_{\alpha,\sigma} a_{\alpha,\sigma}^\dagger a_{\alpha,\sigma} + \sum_{k} \sum_{\sigma = \uparrow, \downarrow} \epsilon_{k,\sigma} b_{k,\sigma}^\dagger b_{k,\sigma} $$

(D4)
and the tunnelling Hamiltonian reads
\[ H_T = t \sum_{\sigma} \left( a_{1,\sigma}^\dagger a_{2,\sigma} + a_{2,\sigma}^\dagger a_{3,\sigma} + \text{H.c.} \right) + t \sum_{k,\sigma} \left( a_{1,\sigma}^\dagger b_{k,\sigma} + b_{k,\sigma}^\dagger a_{3,\sigma} + \text{H.c.} \right). \] (D5)

In Eq. (D4), \( a_{\alpha,\sigma} \) and \( b_{\kappa,\sigma} \) annihiliate electrons with spin \( \sigma \) in the qubit level of dot \( \alpha \) and in the valence band of dot 2, respectively. We assume that \( E_{1,\sigma} = E_{3,\sigma} \) (i.e. qubit 1 and qubit 3 are on resonance) and \( \Delta E_1 = E_{2,\sigma} - E_{1,\sigma} \approx E_{\text{gap}} = 2\hbar v_0 \) (see Fig. 6). In Eq. (D5), we make the approximation that all tunnelling matrix elements \( t \) depend only very weakly on energy and are real. The transmission rate from an initial state \( |i\rangle \) to a final state \( |f\rangle \) can be calculated using Fermi’s golden rule
\[ W_{fi} = \frac{\pi}{\hbar} |\langle f|T(E_i)|i\rangle|^2 \delta(E_f - E_i) \] (D6)

with the transition matrix given by (up to second order in \( H_T \))
\[ T(E) = H_T + H_T^\dagger - \frac{1}{E + i\eta - H_0} H_T + \ldots \] (D7)

We can put \( \eta = 0 \) in the latter equation because we are only interested in off-resonant cotunnelling processes. This means that \( \Delta E_2 \) (see Fig. 6) should be finite because we want to have well-localized qubit states. The corresponding matrix elements of the T-matrix (D7) may be written as
\[ (T(E))_{k,k'} = (H_T)_{k,k'} + \sum_{k''} \left( (H_T)_{k,k''} \frac{1}{E - E_{k''}} (H_T)_{k'',k'} + \ldots \right) \] (D8)

Now, we want to calculate \( |T_{13}|^2 \equiv |\langle 3|T(E_{1,\sigma})|1\rangle|^2 \), where \( |1\rangle \) and \( |3\rangle \) are the ground states of qubit 1 and 3, respectively. The lowest non-vanishing contribution of \( |T_{13}|^2 \) is of cotunnelling type, i.e., of fourth order in \( t \). It is possible to separate the different contributions to \( |T_{13}|^2 \) into three terms, namely
\[ |T_{13}|^2 = |T_{13}^{QB}|^2 + |T_{13}^{VB}|^2 + 2 \text{Re} \left[ (T_{13}^{QB})^\dagger T_{13}^{VB} \right]. \] (D9)

In the latter equation, \( |T_{13}^{QB}|^2 \) determines the transition rate via the qubit level of dot 2 (the unwanted process), \( |T_{13}^{VB}|^2 \) determines the transition rate via the continuum of states in the valence band of dot 2 (the wanted process), and \( 2 \text{Re} \left[ (T_{13}^{QB})^\dagger T_{13}^{VB} \right] \) is the interference term of the two paths. It is straightforward to derive that (for a given spin \( \sigma \) of qubit 2)
\[ T_{13}^{QB}(E_{1,\sigma}) = \frac{t^2}{E_{1,\sigma} - E_{2,\sigma}}. \] (D10)

This has to be compared with
\[ T_{13}^{VB}(E_{1,\sigma}) = \frac{t^2}{\Delta E_{\text{gap}}} \int_{E_{\text{gap}}/2}^{\Delta} dE \frac{\nu_0(E)}{E_{1,\sigma} - E - E}, \]
\[ \approx \frac{L^2}{\pi \hbar} \ln(4\Delta/E_{\text{gap}}), \] (D11)

where
\[ \nu_0(E) = \frac{L}{\hbar \nu \pi} \frac{E}{\sqrt{E^2 - (E_{\text{gap}}/2)^2}} \] (D12)

is the density of states of the mode \( n = 0 \) with \( E_{\text{gap}} = 2\hbar v_0 \).

In Eq. (D11), we integrate over the whole band width of the valence band (bounded by \( \Delta \approx 6 \text{eV} \)). The approximate result in the second line of Eq. (D11) holds for the hierarchy of energies \( \Delta \gg E_{\text{gap}} \gg E_{1,\sigma} \). (In a more general case, the integral in Eq. (D11) can still be evaluated analytically but yields a less compact expression.)

The contribution coming from \( |T_{13}^{QB}|^2 \) is evidently the smallest of the three terms on the rhs of Eq. (D9). If we want to compare the rate that does not affect the spin of the qubit level in dot 2 (which we call \( \Gamma_{VB} \)) with the largest of the rates that does affect the spin of the qubit level in dot 2 (which we call \( \Gamma_{QB} \)), we can estimate
\[ \frac{\Gamma_{VB}}{\Gamma_{QB}} = \frac{|T_{13}^{VB}|^2}{2 \text{Re} \left[ (T_{13}^{QB})^\dagger T_{13}^{VB} \right]} \approx \frac{L}{W} \ln(4\Delta/E_{\text{gap}}). \] (D13)

The latter result is Eq. (9). It shows that by increasing the aspect ratio \( L/W \) we can increase the weight of the coupling via the valence band states of dot 2 (which is wanted) as compared to the weight of the coupling via the qubit state of dot 2 (which is unwanted).

**APPENDIX E: DECOHERENCE**

Finally, we give some arguments and rough estimates for the spin decoherence in graphene. It is generally believed that spin-orbit effects are weaker in carbon than in GaAs due to the lower atomic weight. Therefore, the dominating mechanism for decoherence will be the hyperfine coupling to the nuclear spins that are present in the material.

The coherence time given by the hyperfine coupling can be estimated as \[ t \approx \frac{\sqrt{N}}{cA^{-1}} \], where \( N \) is the number of atoms in the dot (for a typical graphene dot, \( N \approx 10^3 \)), \( cN \) is the number of atoms per dot with a nuclear spin, and \( A \) the coupling constant of the hyperfine interaction. Since we mainly deal with \( \pi \) orbitals in graphene, the contact hyperfine interaction is strongly reduced and the hyperfine interaction is dominated by its dipolar part. From the best available calculations for the dipolar hyperfine matrix elements [32, 33], one obtains
\[ A_{\text{dip}} = 8 \frac{\mu_0}{\Delta r_nodes^2} \mu_B \mu B \left( \frac{1}{\gamma} \right) \approx 0.38 \mu\text{eV}, \] (E1)

where \( \mu \) is the nuclear magneton of \(^{13}\text{C} \), \( \mu_B \) is the Bohr magneton, and \( \mu_0 \) the vacuum dielectric constant. This estimated value for \( A_{\text{dip}} \) is smaller than \( A_{\text{GaAs}} \approx 90 \mu\text{eV} \) by more than two orders of magnitude. The natural abundance of \(^{13}\text{C} \) is about \( c \approx 1\% \) which yields, with the values quoted above, a coherence time of approximately \( t \approx 10 \mu\text{s} \), about a thousand times longer than in GaAs. Unlike in GaAs, this value can be...
improved by isotopic purification. Reducing the $^{13}$C content by a factor of about 100 already decreases the average number of nuclear spins per dot to about one. This allows for a preselection of the dots without any nuclear spin to be used as qubits.


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