An On-Demand Coherent Single-Electron Source

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We report on the electron analog of the single-photon gun. On-demand single-electron injection in a quantum conductor was obtained using a quantum dot connected to the conductor via a tunnel barrier. Electron emission was triggered by the application of a potential step that compensated for the dot-charging energy. Depending on the barrier transparency, the quantum emission time ranged from 0.1 to 10 nanoseconds. The single-electron source should prove useful for the use of quantum bits in ballistic conductors. Additionally, periodic sequences of single-electron emission and absorption generate a quantized alternating current.

In quantum optics, a single-photon source is an essential building block for the manipulation of the smallest amount of information coded by a quantum state: a qubit (1, 2). Combined with beam splitters, polarizers, and projective measurements, several photonic qubits can be manipulated to process quantum information (3). The most celebrated case is the secure transmission of information by means of quantum cryptography. Similarly, one expects that electrons propagating ballistically in ultra-pure low-dimensional conductors can perform quantum logic tasks in perfect analogy with photons propagating in optical media (4–6). The analogy has a long history (7) and has provided illuminating comparisons between the intensity of light and that of electrical current; between photon noise and electrical shot noise (8, 9); and, more recently, between photon and electron quantum entanglement (10–12). Because electrons are fermions, entanglement offers new routes not possible with photons (12). Practically, electronic analogs of beam splitters and Fabry-Pérot and Mach-Zehnder interferometers (13, 14) have been used in ballistic conductors, providing the necessary quantum gate for an all-linear electron optics quantum computation. Yet missing were a single-electron source and a single-electron detector (15) suitable for coherent emission and projective measurements. The former initializes quantum states, whereas the latter reads the final states after electrons have passed through the quantum gates.

Unlike the case of photons, the creation of single-electron sources is expected to be simpler because of Fermi statistics and Coulomb interaction. For example, considering a voltage-biased single-mode conductor, a contact at energy $eV'$ above the energy of the other contact is known to inject single electrons into the conductor at a regular rate $eV'/h$, thereby leading to the quantization of dc conductance in quantum point contacts (QPCs) (16, 17). A second example is the electron pump, in which a dc current is produced by sequential time-controlled transfer of single electrons between metallic islands in series (18, 19) or by the manipulation of tunnel barriers of quantum dots (20, 21). The cost in Coulomb charging energy to add or remove an electron ensures a well-defined electron number in each island or dot. These two sources are, however, not useful for quantum information. In the first case, there is no time control of the electron injection. Because only statistical measurements are possible, the biased contact is suitable for demonstrating coherent phenomena such as interferences or electron entanglement (10, 11) but not for manipulating quantum information. In the second example, time-controlled injection can be realized, but the energy of emitted electrons is expected to spread, at random, in an energy range much larger than the tunneling rate (typically a fraction of the charging energy, depending on the pumping conditions). The statistical distribution in energy will smear the coherent effects required for manipulating the quantum information. Finally, a third approach has been theoretically proposed in (22–24), considering voltage pulses $V(t)$ applied to an ohmic contact. When the Faraday flux $eV(t)/2\pi\hbar$ is an integer, an integer number of electrons is injected. Here noiseless injection requires that the pulses have a special Lorenzian shape and exact integer Faraday flux, otherwise logarithmic divergence of the charge fluctuations occurs. No experiment is available yet to test these ideas.

We report here on the realization of a time-controlled single-electron source suitable for the coherent manipulation of ballistic electronic qubits, which emits the electrons into a well-defined quantum state. The injection scheme is different from those considered above. The source is made of a quantum dot, realized in a two-dimensional (2D) electron gas in gallium arsenide (GaAs) semiconductors and tunnel-coupled to the conductor through a QPC. By application of a sudden voltage step on a capacitively coupled gate, the charging energy is

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compensated for and the electron occupying the highest energy level of the dot is emitted. The final state of the electron is a coherent wave packet propagating away in the conductor. Its energy width is given by the inverse tunneling time, as required for an on-demand single-particle source, and is independent of temperature. Its mean energy can be adjusted above the Fermi energy by tuning the voltage step amplitude. The circuit (Fig. 1A), is realized in a 2D electron gas in a GaAsAl/GaAs heterojunction of nominal density $n_s = 1.7 \times 10^{15}$ m$^{-2}$ and mobility $\mu = 260$ V$^{-1}$ m$^2$ s$^{-1}$. The dot is electrostatically coupled to a metallic top gate, 100 nm above the 2D electron gas, whose ac voltage, $V_{\text{exc}}$, controls the dot potential at the subnanosecond time scale. For all measurements, the electronic temperature is about 200 mK and a magnetic field $B \approx 1.3$ T is applied to the sample so as to work in the quantum Hall regime where no spin degeneracy. The QPC dc gate voltage $V_g$ is tuned to control the transmission $D$ of a single edge state as well as the dc dot potential. As reported (25), this circuit constitutes the paradigm of a quantum-coherent resistor-capacitor (RC) circuit, where coherence is seen to strongly affect the charge relaxation dynamics. From this study, the charging energy $\Delta + e^2/\text{C} \approx \Delta \approx 2.5$ K was extracted (26). Here the large top-gate capacitance makes the Coulomb energy $e^2/\text{C}$ unusually small, and the total charging energy reduces to the energy-level spacing $\Delta$.

In (25), the linear response of the current to the ac top-gate voltage was investigated, and the ac charge amplitude was much lower than the elementary charge $e$. In order to achieve single-charge injection, we had to apply a high-amplitude excitation ($V_{\text{exc}} \sim \Delta/e$) and go beyond the linear regime. When an electron is suddenly brought above the Fermi energy of the lead, it is expected to escape the dot at a typical tunnel rate $\tau^{-1} = D\Delta/h$, where $\Delta/h$ is the attempt frequency and $D$ is the transmission probability. This gives nanosecond time scales, for which single-charge detection is still out of reach experimentally. To increase the signal-to-noise ratio, a statistical average over many individual events is used by generating repetitive sequences of single-electron emission followed by single-electron absorption (or hole emission) as shown in Fig. 1A. This is done by applying a periodic square wave voltage amplitude $\approx \Delta/e$ to the top gate. Figure 1B shows typical temporal traces of the current averaged over a few seconds for a repetition period of $T = 32$ ns. The single-electron events reconstruct the exponential current decay of an RC circuit. When transmission $D$ is decreased from $=0.03$ to $=0.002$, the relaxation time $\tau$, extracted from the exponential decay, increases from 0.9 to 10 ns. For the two highest transmissions in Fig. 1B, $\tau \ll T/2$, the currents decay to zero, and the mean transferred charge per half period is constant. For the smallest transmission, $\tau \sim T/2$, the mean emitted charge decreases because electrons have reduced probability of escaping the dot. These time-domain measurements are limited by the 1-GHz bandwidth of the acquisition card and give access to the few-nanosecond injection times corresponding to small transmissions $D \lesssim 0.03$.

In order to get a better understanding of the above results, we extend the harmonic linear re-

![Fig. 1. Single-charge injection. (A) Schematic of single-charge injection. Starting from an antiresonant situation where the Fermi energy lies between two energy levels of the dot (step 1), the dot potential is increased by $\Delta$ moving one occupied level above the Fermi energy (step 2). One electron then escapes the dot on the mean time $\tau = h/D\Delta$. The dot potential is then brought back to its initial value (step 3), where one electron can enter it, leaving a hole in the Fermi sea. (Inset at right) The quantum RC circuit: one edge channel is transmitted inside the submicrometer dot, with transmission $D$ tuned by the QPC gate voltage $V_g$. The dot potential is varied by a radio-frequency excitation $V_{\text{exc}}$ applied on a macroscopic gate located on top of the dot. The electrostatic potential can also be tuned by $V_g$ because of the electrostatic coupling between the dot and the QPC. (B) Time-domain measurement of the average current (black curves) on one period of the excitation signal (red curves) at $2eV_{\text{exc}} = \Delta$ for three values of the transmission $D$. The relaxation time $\tau$ is deduced from an exponential fit (blue curve).

![Fig. 2. $I_\omega$ as a function of $V_g$ at $f = 180$ MHz for different values of the excitation amplitude $2eV_{\text{exc}}$. Transmission $D$ is also indicated. (A) Modulus $|I_\omega|$. The horizontal dashed line is the constant value $|I_\omega| = 2ef$. (B) Nyquist representation (imaginary part of $I_\omega$ versus real part of $I_\omega$). The red curve corresponds to an RC circuit of constant capacitance $e^2/\text{C}$ and varying resistance. (C) Phase $\phi$. The phase $\phi$ is independent of $V_{\text{exc}}$.](image-url)
sponse theory of a quantum RC circuit (27–29) to calculate the nonlinear response to a high-amplitude square excitation voltage ($eV_{\text{exc}} > hf$).

Calculation shows that the circuit still behaves as an RC circuit with a current given by

$$I(t) = \frac{q}{\tau} e^{-t/\tau} \text{ for } 0 \leq \tau \leq T/2$$

where $N(e)$ is the dot density of states and $f(e)$ denotes the Fermi-Dirac distribution. The nonlinear capacitance and charge relaxation resistance can be defined respectively by $C_q = q/2eV_{\text{exc}}$ and $R_q = \tau/C_q$. For unit transmission $D = 1$, electrons are fully delocalized, $N(e)$ is uniform, and the charge $q$ evolves linearly with $V_{\text{exc}}$ as expected. In contrast, for low transmission, $N(e)$ is sharply peaked on well-resolved energy levels, and $q$ exhibits a staircase dependence on $V_{\text{exc}}$ with steep steps whenever one electronic level is brought above the Fermi energy. Thus, our calculations establish the process of single-electron injection depicted in Fig. 1. For a dot energy spectrum with constant level spacing $\Delta$, a remarkable situation occurs when $2eV_{\text{exc}} = \Delta$, as $q = e$ and $C_q = e^2/\Delta$ irrespective of the transmission $D$ and of the dc dot potential. As a matter of fact, Eq. 2 shows that, in these conditions, $q$ is given by integrating $N(e)$ over exactly one level spacing. For $D \ll 1$, we recover the Landauer formula for the resistance $R_q = h/2e^2$, and the escape time is given by $\tau = h/\Delta$, as expected from a semiclassical approach. The exponential current decay, the constant injection charge for $\tau < T/2$, and the decrease of $\tau$ with transmission $D$ account well for our experimental observations in Fig. 1B.

For a more accurate experimental determination of $q$ and $\tau$ and to investigate subnanosecond time scales, we consider in the following measurements the current first harmonic $I_{\omega}$ at higher frequencies $f = \omega/2\pi = 1/T$. Following Eq. 1, we have

$$I_{\omega} = \frac{2qf}{1 - i\omega\tau}$$

so that the modulus $|I_{\omega}|$ and the phase $\phi = \arctan(\omega\tau)$ allow for the determination of $q$ and $\tau$.

Figure 2A shows $|I_{\omega}|$ measured as a function of QPC gate voltage $V_G$ at $f = 180$ MHz for increasing values of the excitation voltage $2eV_{\text{exc}}$. The range of $V_G$ maps the full transmission excision $D = 0$ to 1. The low-excitation $2eV_{\text{exc}} = \Delta/4$ data nearly correspond to the linear response reported in (25). The current exhibits strong oscillations reflecting the variation with $V_G$ of the dot density of states at the Fermi energy. At larger excitation voltages, the current peaks are broadened as expected from Eq. 2 when $2eV_{\text{exc}}$ gets larger than thermal energy ($k_B T$). For $2eV_{\text{exc}} = \Delta$, the oscillations disappear completely and $|I_{\omega}| = 2\text{ef}$, down to a low transmission threshold $D \sim 0.05$. The oscillations reappear for larger excitations. The constant current $|I_{\omega}| = 2\text{ef}$ is the frequency-domain counterpart of the constant charge regime observed in the time domain, for the injection and absorption of a single electron per half period. The cutoff observed for $D \leq 0.02$ corresponds to the limit $\omega\tau \gtrsim 1$, where the escape time $\tau$ exceeds $T/2$. The constant $C_q$ regime obtained for $2eV_{\text{exc}} = \Delta$ can be viewed in the Nyquist representation of Fig. 2B. The corresponding diagram is the half-circle that is characteristic of an RC circuit with a constant capacitance $e^2/\Delta$ and transmission-dependent resistance. In contrast, the curves obtained for larger or smaller excitations exhibit strong capacitance oscillations.

Figure 2C represents the phase $\phi = \arctan(\omega\tau)$ of the current as a function of $V_G$ for different excitation voltages. $\phi$ shows a quasi-monotonic $\pi/2$ sweep in increasing transmission. The absence of residual oscillations proves that $\tau$ is nearly insensitive to the dot potential. As seen in Fig. 2C, $\tau$ is also independent of $V_{\text{exc}}$. In Fig. 3, we have gathered the values of $I_{\omega}$ obtained from 1-GHz bandwidth time-domain measurements at a 31.25-MHz repetition rate and from frequency-domain measurements at 180 and 515 MHz. The measurements probe a very broad transmission range ($D = 0.002$ to 0.2) corresponding to escape times varying from 10 ns to 100 ps. In the overlapping range, the different independent determinations coincide within error bars, agreeing quantitatively with the prediction $\tau = h/\Delta$ also represented in Fig. 3, where the dependence $D(V_G)$ is deduced from the linear regime (25).

We now discuss the conditions for single-electron injection leading to a good quantization of the ac current as a figure of merit of single-charge injection. Figure 4A represents $|I_{\omega}|$ as function of $V_{\text{exc}}$ for typical values of the dc dot potential at fixed transmissions $D \approx 0.2$ and
The Catalytic Cross-Coupling of Unactivated Arenes

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The industrially important coupling of aromatic compounds has generally required differential preactivation of the arene coupling partners with a halide and an electropositive group. Here we report that palladium, in conjunction with a copper oxidant, can catalyze the cross-coupling of N-acetyllindoles and benzenes in high yield and high regioselectivity across a range of indoles without recourse to activating groups. These reactions are completely selective for arene cross-coupling, with no products arising from indole or benzene homo-coupling detected by spectroscopic analysis. This efficient reactivity should be useful in the design of other oxidative arene cross-couplings as well.

The immense scientific and commercial value of biaryl molecules is illustrated by their ubiquity as building blocks in light-emitting diodes, electron transport devices, liquid crystals, and medicinal compounds (1). The structural simplicity of biaryl compounds belies their preparative complexity, and the search for efficient and convergent syntheses has captivated the attention of synthetic chemists for more than a century. Over the past 30 years, biaryl cross-coupling reactions based on carbon fragment preactivation have revolutionized our ability to forge the carbon-carbon biaryl linkage (1, 2). Of these reactions, the most widely accepted and used are the palladium-catalyzed cross-coupling reactions (such as the Suzuki reaction) of aryl halides and aryl organometallics (3). As is common today, these reactions are dependent on preactivation of the two aromatic carbon fragments with halides and electropositive groups, such as boronic acids or stannanes (4). Incorporation of these functional groups can require several synthetic steps,