Observation of a Nanoscale Metallic Dot Self-Consistently Coupled to a Two-Level System

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Abstract

We have observed anomalous transport properties for a 50 nm Bi dot in the Coulomb-blockade regime. Over a range of gate voltages, Coulomb blockade peaks are suppressed at low bias, and dramatic structure appears in the current at higher bias. We propose that the state of the dot is determined self-consistently with the state of a nearby two-level system (TLS) to which it is electrostatically coupled. As a gate voltage is swept, the ground state alternates between states of the TLS, leading to skipped Coulomb-blockade peaks at low bias. At a fixed gate voltage and high bias, transport may occur through a cascade of excited states connected by the dynamic switching of the TLS.

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With the Coulomb blockade in single-electron transistors [1,2] and quantum dots [3,4] now firmly established, efforts have begun to turn towards more complex systems in which these artificial atoms form the building blocks. Significant progress has been made in understanding linear-response transport in what may be termed “artificial molecules” in studies of double quantum dots [5,6], as well as “artificial solids” in studies of arrays of quantum dots [7,8]. Moreover, such multi-dot systems have been considered as the basis for novel computer memory and logic elements [9]. In these applications the transport properties must be considered well beyond the regime of linear-response.

In this Letter, we examine the transport properties of a quantum dot coupled to a single two-level system (TLS) as a model for nonlinear transport phenomena in systems containing artificial atoms. We report a set of anomalous transport properties of a small Bi dot in the Coulomb-blockade regime. The anomalies are explained by the presence of a TLS in close proximity to the dot. While TLS’s have been studied as a source of $1/f$ noise in MOSFET’s [10,11], quantum point contacts [10,12], and single-electron transistors (SET’s) [13,14], exhibited as random telegraph signals, the coupling in our devices leads to non-stochastic configurations of the TLS. At low bias, reversible switching of the TLS leads to skipped Coulomb blockade peaks in a manner similar to peak suppression in double quantum dots [15]. At higher bias, a novel effect emerges in which transport occurs through a cascade of excited states which are connected by the dynamic switching of the TLS. Similar effects may be expected in multi-dot systems.

The devices, shown schematically in the inset to Fig. 1, are SET’s in the standard double-junction geometry with a capacitively coupled gate [1,2]. They were fabricated using e-beam lithography to make a shadow-mask for use with the self-aligned double-angle evaporation technique [16]. The leads, consisting of Cu wires 20 nm in diameter, were deposited first. They were then oxidized in-situ for 30 minutes at an oxygen pressure of $1 \times 10^{-4}$ Torr before deposition of the Bi dot. The resultant junction resistances of $R_J \geq 1$ MΩ satisfy the condition for the Coulomb blockade $R_J > R_q = \hbar/4e^2 \approx 6.5$ kΩ. Voltage-biased DC measurements were performed in an Oxford dilution refrigerator held at the base temperature of 70 mK.

In the measured devices, the diameter of the dot is roughly 50 nm. As the size of the Bi dot is smaller than a typical grain in a film of Bi grown on the same surface, the dot is likely a single crystal. Furthermore, while quench-condensed amorphous Bi is superconducting, crystalline Bi is not. Bulk crystalline Bi is a semimetal, with a low Fermi energy, $E_F = 27.2$ meV, and a low electron density, $n = 2.7 \times 10^{17}/\text{cm}^3$ [17], about $10^5$ times lower than a typical metal. A small overlap of bands at the Fermi level leads to an equal number of electrons and holes. The highly anisotropic Fermi surface results in Fermi wavelengths $\lambda_F = 14–215$ nm. These wavelengths are comparable to the dimensions of the dot, and quantum properties might be expected [3,18]. However, the devices reported here exhibited only metallic behavior.

Experiments were also conducted using Al as the dot material. The five Al devices studied exhibited only the standard Coulomb-blockade behavior. The anomalous behavior presented here occurred in all seventeen of the Bi dots studied.

The most striking feature of the data is seen in IV curves presented as a function of both bias and gate voltages, $V_b$ and $V_g$, respectively. In a series of $IV_b$ curves taken over a full period of the Coulomb blockade in $V_g$, an anomalous gap remains where the blockade
would normally vanish (Fig. 1). This can also be seen in $IV_g$ sweeps in the same region (Fig. 2(a)), where at low bias ($V_b \ll E_c/e$, where $E_c$ is the charging energy of the dot), the conductance peaks disappear. The range of gate voltages in which the suppression occurred varied from device to device, as did the number of missing peaks, with a maximum of about five. Each device exhibited only one region of peak suppression within the range of gate voltages studied, which corresponded to changing the number of electrons on the dot by up to 100.

At higher biases, where $V_b$ is greater than the anomalous gap, but still smaller than $E_c/e$, the missing peaks emerge and typically split, as seen in Fig. 2(a). At even higher biases, the peaks are suppressed within an envelope, and the lineshapes become complex. An example of this from a different device is shown in Fig. 3. A notable feature of this device is the noise in the peaks, visible even at the measurement bandwidth of 1 Hz. The noise has a broad spectrum, as evidenced in measurements with a bandwidth of 1 kHz (Fig. 3, left inset).

Before introducing the TLS, we first consider other possible origins of these anomalies. A gap similar in appearance to Fig. 1 may arise in superconductors [19]. This would require that both the dot and the leads are superconducting. While Bi in its amorphous state may be superconducting, the Cu leads are not. Moreover, a magnetic field up to 9 T does not remove the anomalous gaps. The opening of a gap at the band overlap in the Bi semimetal due to size quantization may also be considered [20], but such a bandgap would not manifest itself as in Fig. 1. The result of a bandgap $E_g$ in the Bi dot would be to increase the spacing in gate voltage $\Delta V_g$ between the conductance peaks which occur at the band edges [4]:

$$\Delta V_g = \frac{C_\Sigma}{eC_g} (E_c + E_g) = \frac{e}{C_g} + \frac{C_\Sigma}{C_g} \frac{E_g}{e}$$

(0.1)

where $E_c = e^2/C_\Sigma$ is the charging energy, and $C_g$ and $C_\Sigma$ are, respectively, the capacitance of the dot to the gate and the total capacitance of the dot. Finally, we may exclude migrating charges as the data are highly reproducible.

In what follows, we show that the transport anomalies may be explained by postulating a TLS coupled electrostatically to the Bi dot. We imagine the TLS to consist of a double-well potential (Fig. 1 inset) in which a charged particle is free to move. Electrons on the dot interact with the charge in the TLS. Hence a change in the number of electrons on the dot may change the relative energies of the two states of the TLS. In turn, the switching of the TLS will act as an effective change in gate voltage for the dot. Thus, the charge state of the dot must be determined self-consistently with the state of the TLS.

To model in detail the behavior of a dot electrostatically coupled to a two-level system, we consider the following classical Hamiltonian:

$$H_{ON/OFF} = \frac{1}{2C_\Sigma} (Q - Q_0 - Q_{0_{ON/OFF}}^2) + V_g Q_{TLS}^{ON/OFF} + E_{TLS}^{ON/OFF}$$

(0.2)

where ON/OFF denotes the state of the TLS. Without the terms due to the two-level system, this is just the ordinary Hamiltonian of a metal dot. The actual integer charge on the dot is given by $Q$, while the optimal charge is controlled by the gate voltage $Q_0 \propto V_g C_g$. The electrostatic coupling between the TLS and the dot can be expressed by a shift $Q_{0_{ON/OFF}}$ of the optimal charge on the dot. The TLS is also electrostatically coupled to the gate, yielding the linear term $V_g Q_{TLS}^{ON/OFF}$. Lastly, there may be an internal energy difference between the states.
of the TLS, represented by $E_{\text{TLS}}^{\text{ON/OFF}}$. The model is also applicable to multilevel systems, in which case the TLS parameters can take on several discrete values.

By an appropriate choice of $Q_0$, we may set the OFF-state parameters to be zero. This choice for the Hamiltonian leads to the two families of parabolas shown in Fig. 2(b). The horizontal offset, vertical offset, and slope of the ON-state parabolas are determined by $Q_0^{\text{ON}}$, $E_{\text{TLS}}^{\text{ON}}$, and $Q_{\text{TLS}}^{\text{ON}}$, respectively.

As shown in Fig. 2(b), as the gate voltage is increased, the lowest energy configuration may switch back and forth many times between the TLS OFF and ON states. This provides a natural explanation for the missing Coulomb-blockade peaks at low bias voltages in Fig. 2(a). For example, the first switching ON of the TLS occurs together with a switch from $N$ to $N + 1$ electrons on the dot (first dark circle in Fig 2(b)). For this double switch to occur, either the electron number must first change with the TLS fixed, or the TLS must first switch with the electron number fixed. Either of these processes alone requires activation energy. Hence the double switch has an activation barrier and is too slow to produce a measurable current [21]. This process preempts the usual Coulomb-blockade peak which would have occurred at slightly higher gate voltage. The peaks continue to be suppressed at low bias until increasing $V_g$ causes the TLS to switch permanently ON, at which point conductance peaks resume. The number of missing peaks can be estimated from Eq. (0.2) as [22]

$$\frac{e}{|Q_{\text{TLS}}^{\text{ON}}|} \text{Mod} \left( \frac{|Q_0^{\text{ON}}|}{e} \right) \left[1 - \text{Mod} \left( \frac{|Q_0^{\text{ON}}|}{e} \right) \right],$$

such that a small slope $|Q_{\text{TLS}}^{\text{ON}}|$ can lead to a large number of missing peaks.

To understand the more complex behavior at higher bias voltages, we have simulated transport through the dot using the orthodox Coulomb-blockade model [2], but including the TLS. We consider the transport current between a right lead with voltage $V_b/2$ and a left lead with voltage $-V_b/2$, both leads coupled via equal junction resistances $R$ to the dot [23]. Assuming rapid thermalization of electrons on the dot, we solve the rate equations for transitions between different charge states of the dot. However, we also allow transitions to occur between the two states of the TLS. The transition rate is $\Gamma_{\text{TLS}}$ if the overall energy is thereby lowered, and $\Gamma_{\text{TLS}} \exp(-\Delta E/k_B T)$ if the overall energy is raised by $\Delta E$.

As shown in Figs. 2(a) and 3, the simulated current is similar to that observed experimentally, remarkably so in the latter. In both cases, the fitting parameters are those of the TLS (Eq. [1]). In Fig. 2(a), the best fit was obtained in the saturated limit of $\Gamma_{\text{TLS}}$ much faster than the electron tunneling rate, but the opposite limit was obtained in Fig. 3. Thus there appears to be a considerable variation of TLS tunneling rates from device to device.

The interactions between the dot and the TLS may lead to a novel transport process in which the TLS is active, switching dynamically. Consider the inset of Fig. 2(b), which shows the states of the system and the simulated current. There are four accessible states A - D ($V_b > 0.2E_c$), with $N$ or $N + 1$ electrons on the dot, and with the TLS either ON or OFF. At this gate voltage, the bias voltage is sufficient to add an electron to the dot, going from state A to B, with the TLS OFF. The extra electron may escape into the other lead, causing relaxation from B back to A. However, it is also possible for the TLS to spontaneously switch from OFF to ON, since $\Delta E$ is negative for this process, thereby taking the system from state B to state C. Now, the system may switch repeatedly between states C and
**D** resulting in electron transport, with the TLS staying in the ON state. Alternatively, when the system is in state **D** with \( N \) electrons, the TLS may switch back to OFF without changing the number of electrons on the dot, and the system is back where it started in state **A**. This process can then begin again, each cycle transferring a minimum of two electrons through the dot.

If the TLS transition rate \( \Gamma_{\text{TLS}} \) is large compared with the tunnelling rate, then these cycles, where each successive electron moving through the dot is accompanied by a switch of the TLS, will dominate the transport. If the TLS is slow, then the system will switch telegraphically back and forth between transport with the TLS ON or OFF, each process exhibiting a different conductance. This may explain the noise seen in the device of Fig. 3.

While it is evident that the TLS is associated with Bi, since these effects were not observed in our Al dots, the microscopic origin of the TLS is unknown. It may reside on the surface of the dot in the Bi oxide layer, or it may be due to an ancillary grain of Bi from the double-angle evaporation. If it were a bistable defect, the associated small charge displacement, on the order of a lattice spacing, would be an insignificant effect in our device geometry. A larger effective charge displacement may come from a trapping state which is either filled or unfilled [14]. The dynamics of such a charge trap may then be responsible for the broad range of lifetimes observed.

The phenomenology of the dot-TLS system may be important in a number of applications. In quantum dots, a system consisting of two dots connected in parallel [24] will mimic a dot coupled to a two-level system if one of the dots is electrically coupled to only one lead. With perhaps broader implications, a single-electron transistor used as an electrometer [25] may behave in a way similar to the dot-TLS system. Such an electrometer, which may be used to detect charge configurations, *interacts with the system being measured*. For example, the Bi dot in our devices may be said to measure the state of the TLS, but clearly this process sometimes changes the state of the TLS. Therefore, in using an electrometer to probe a system of mobile charges, additional movement of the charges due to the measurement process must also be considered.

In conclusion, we have observed anomalous transport characteristics in a 50 nm Bi dot, including suppression of Coulomb-blockade conductance peaks at low bias voltages, and splitting of these peaks at higher bias. This behavior may be ascribed to an electrostatic coupling of the dot to a TLS. The interaction with the dot causes the TLS to switch back and forth multiple times as the gate voltage is increased. Furthermore, sufficient bias may result in a transport process where the TLS switches dynamically with transport of each electron through the dot. A similar effect is expected in some double-dot systems. It may be informative to study the dynamics of such dot-TLS systems, particularly if the switching rate of the TLS can be controlled. Finally, more complicated dynamics may be expected in dots coupled to multilevel systems.
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FIGURES

FIG. 1. $I$ vs. $V_b$ curves as a function of gate voltage $V_g$, over a full period of the Coulomb blockade corresponding to the region of peak marked with an arrow in Fig. 2(a). An anomalous gap occurs where the current is expected to be nearly linear, as indicated by the dotted line. Inset: Schematic of the devices, consisting of a Bi dot coupled to two Cu leads via tunnel junctions, with a capacitively coupled gate. Nearby is the inferred two-level system (TLS) containing a single charge which is electrostatically coupled to the dot.

FIG. 2. A series of $I$ vs. $V_g$ sweeps in (a) for bias voltages $V_b > 0$, $T = 70$ mK. Offset for clarity, the flat regions mark $I = 0$ for each curve. The top curve is the simulated current, for $V_b = 4$ mV, $E_c/e = 8.2$ mV and $T = 70$ mK as in the top experimental curve, and fitted two-level system (TLS) parameters (Eq. 0.2) $Q^{0\text{ON}}_0/e = 0.23$, $Q^{0\text{ON}}_{\text{TLS}}/e = -0.1$, $E^{0\text{ON}}_{\text{TLS}}/E_c = 0.39$, and $\Gamma^{0\text{ON}}_{\text{TLS}} = 100hE_c/e^2R$. The rounding of the data relative to the simulation suggests that the temperature of the dot electrons is higher than 70 mK. The energy of the dot-plus-TLS system is shown in (b) for both states of the TLS (solid line=OFF, dashed line=ON). $N$ indicates the number of electrons on the dot. For low bias $V_b \ll E_c/e$, the system will follow the lowest energy curve, switching at the crossover points indicated (dark circles), preempting the crossovers associated with Coulomb-blockade peaks. Inset: Expanded view of the states relevant to the peak marked with an arrow in (a), shown with the simulated current. At higher bias, transport will take place through the sequence of states A-D where the TLS switches with each electron passing through the dot, as described in the text.

FIG. 3. Effect of the TLS switching at a bias $V_b \simeq E_c/e$ for a different device than in Fig. 2 (solid line). The simulated current (dotted line) reproduces many features of the data with fitted TLS parameters $Q^{0\text{ON}}_0/e = -0.3$, $Q^{0\text{ON}}_{\text{TLS}}/e = -0.048$, $E^{0\text{ON}}_{\text{TLS}}/E_c = 0.27$, and $\Gamma^{0\text{ON}}_{\text{TLS}}/e = 0.01hE_c/e^2R$, and the experimental parameters $V_b = 0.9$ mV, $E_c/e = 1.5$ mV, $T = 140$ mK, and bandwidth=1 Hz. The temperature used in the fit was $T = 345$ mK, indicating that the small, isolated grain of Bi may not cool to the temperature of the leads. Left inset: Expanded view of peak indicated by arrow, with bandwidth=1 kHz. Right inset: Simulated current shown by itself for clarity.
Fig. 1
Fig. 2
Fig. 3