Electrically Tunable Spin Polarization in a Carbon Nanotube Spin Diode

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We have studied the current through a carbon-nanotube quantum dot with one ferromagnetic and one normal-metal lead. For the values of gate voltage at which the normal lead is resonant with the single available nondegenerate energy level on the dot, we observe a pronounced decrease in the current for one bias direction. We show that this rectification is spin dependent, and that it stems from the interplay between the spin accumulation and the Coulomb blockade on the quantum dot. The degree of resulting spin polarization is fully and precisely tunable using the gate and bias voltages.

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The ability to create, manipulate, and detect spin currents is central to the development of spintronic devices [\[1\]](#page-2-0). Controlling the spin currents by purely electrical means [\[2](#page-2-1),[3](#page-3-0)] is particularly interesting, as that would allow the integration of spintronic devices with conventional electronics. Creating spin-polarized currents typically involves injection from ferromagnets [[4](#page-3-1)[,5\]](#page-3-2) or magnetic semiconductors [[6](#page-3-3),[7](#page-3-4)]. Spin filters, spin memory devices and spin pumps using quantum dots with normal-metal leads have also been proposed $[8,9]$ $[8,9]$ $[8,9]$. If a quantum dot is connected to two ferromagnetic leads with different polarizations, it can operate as a spin-diode, as predicted recently $[8,10-14]$ $[8,10-14]$ $[8,10-14]$. In this system, the spin dependent asymmetry in the tunneling rates between the two junctions is predicted to lead to spin accumulation, current rectification and spinpolarized currents. If a quantum dot is attached to one ferromagnetic and one normal lead, the effect is even more pronounced $[15-17]$ $[15-17]$ $[15-17]$ $[15-17]$ and can potentially be used for spin injection [\[18\]](#page-3-11). The current polarization in such a system can be controlled entirely by the gate and bias voltages. In this Letter, we report a pronounced asymmetry in the current-voltage characteristics of a quantum dot with one ferromagnetic and one normal-metal lead. The observed asymmetry is analyzed in terms of spin dependent tunneling rates and the results show excellent agreement with the theoretical predictions. Our device therefore represents an experimental realization of a tunable spin diode.

The sample consists of a carbon-nanotube (CNT) [\[19](#page-3-12)[,20\]](#page-3-13) quantum dot connected to cobalt and niobium electrodes. CNTs are ideal candidates for fabricating spin-based devices because of their long spin-coherence lengths [\[21\]](#page-3-14). They typically form tunnel barriers with metallic contacts so that small nanotube sections contacted by metal electrodes behave like quantum dots at low temperatures [[22](#page-3-15)]. CNTs were grown using chemical vapor deposition [[23](#page-3-16)] and positioned using ac dielectrophoresis [\[24\]](#page-3-17), and the electrodes were fabricated using the standard electron-beam lithography methods. An atomic force microscope image of the sample is shown in Fig. [1\(a\)](#page-1-0). Measurements were performed by applying a bias voltage and measuring the current through the sample. A capacitively coupled gate voltage was applied to the sample through a thermally grown $SiO₂$ layer, as shown in the measurement schematic in Fig. $1(b)$. All measurements were carried out at temperatures of 10 K, above the superconducting transition temperature of niobium. We have studied several samples, with the sections of carbon nanotubes between the leads typically between 250 and 500 nm long. In this work, we present the results obtained from the representative sample shown in Fig. [1\(a\).](#page-1-0)

A map of the two-terminal differential conductance, $G = dI/dV$, is shown as a function of bias and gate voltages in Fig. [2\(a\).](#page-1-1) We observe typical quantum dot behavior characterized by the diamond-shaped regions where the electron transport through the device is forbidden due to the Coulomb blockade [[25](#page-3-18)]. The charging energy and level spacing of the dot are 7 and 3 meV, respectively [\[26\]](#page-3-19). For the range of gate and bias voltages shown in Fig. $2(a)$, the Coulomb diamonds are regular and do not vary in size. In this region, we do not observe the even-odd filling pattern of the quantum dot, typically seen when the energy levels on the dot are spin degenerate [\[27\]](#page-3-20).

The conductance as a function of gate voltage for positive $(G₊)$ and negative $(G₋)$ bias is shown in Fig. [2\(b\).](#page-1-1) The data have been fit using the Breit-Wigner line shape model [\[25\]](#page-3-18), using the capacitance ratio $\alpha = 0.03$ [\[22\]](#page-3-15) and a temperature of 10 K. A pronounced asymmetry between the positive-bias and negative-bias conductance traces is observed in Fig. $2(b)$. In particular, around the gate voltage values of $V_g = 8.45, 8.85,$ and 9.25 V, the peaks in G_{+} are strongly suppressed relative to the corresponding peaks in *G*. The normal-metal lead is resonant for these values of gate voltage.

An asymmetry in conductance with respect to bias direction is typically observed in quantum dots which do not have symmetrically coupled leads. However, we will argue that there is a significant contribution stemming from spindependent suppression of tunneling when electrons are coming from the resonant normal lead.

FIG. 1 (color online). (a) Atomic force microscope image of the sample (scale bar: $1 \mu m$). The distance between the electrodes is 300 nm. CNT height is measured as *<*2 nm. The electrodes are cobalt and niobium, as indicated on the image. (b) Schematic of the measurement setup. Bias is applied between the niobium and the cobalt lead with niobium lead grounded. Gate voltage is applied to the doped silicon substrate through a 500 nm thick thermally grown layer of $SiO₂$.

To highlight this spin dependent contribution to the observed asymmetry, we analyze the single-junction conductances for each of the leads. The single-junction conductances are labeled as $G_{N(FM)}$, where *N* and FM denote the normal metal and the ferromagnet, respectively. An additional subscript $(+ \text{ or } -)$ refers to either positive or negative bias. The single-junction conductances were not measured directly, but were obtained as a part of the Breit-Wigner fitting procedure for the data shown in Fig. [2\(b\)](#page-1-1). Since a normal tunnel junction must be symmetric with respect to bias direction [\[26\]](#page-3-19), we correct for the asymmetry caused by the quantum dot-electrode coupling by scaling the single-junction conductances so that G_{N+} and G_{N-} match. A similar scaling of the ferromagnetic singlejunction conductances shows that they remain asymmetric as seen in Fig. [3\(b\).](#page-2-2) In particular, G_{FM+} is *lower* than G_{FM-} for certain values of gate voltage, which means that the tunneling is suppressed in the case of positive bias as compared to the negative bias. The values of gate voltage for which this suppression occurs correspond to the situations in which the normal lead is resonant with the single energy level available on the dot. Such suppression can only occur if the electrons tunnel more easily from the ferromagnetic lead onto the dot rather than vice versa for those particular values of gate voltages.

To describe the nature of this asymmetry in a more quantitative way, we assign different tunneling rates to the two tunnel junctions: Γ_{FM} refers to the tunneling rate through the ferromagnetic contact, while Γ_N refers to the tunneling rate through the normal contact. Both tunneling rates depend on the available energy levels on the dot, and it is reasonable to assume that Γ_{FM} also depends on the spin polarization of the ferromagnetic electrode. For the ferromagnetic lead we take $\Gamma_{FM} = \Gamma_0(1 \pm p)$ [[11](#page-3-21)], where *p* is the thermodynamic polarization of the ferromagnet, positive (negative) sign is for majority (minority) spin, and Γ_0 is a constant. The normal-metal lead is not spin dependent, so we take the tunneling rate to be $\Gamma_N = \gamma \Gamma_0$ [\[11\]](#page-3-21). The factor γ is the ratio of the single-junction conductances

 $(\gamma = G_N/G_{FM})$, which accounts for the asymmetry in the electrode couplings as a function of the gate voltage.

We start with an expression for the current [\[15\]](#page-3-9) derived by employing the master equation approach [\[28\]](#page-3-22) in the sequential tunneling regime. Using the tunneling rates defined above, we find the total current through a quantum dot with a single ferromagnetic lead to be

$$
I = \begin{cases} 2\Gamma_0 e \gamma \frac{1 - p^2}{(1 - p^2) + 2\gamma} & eV > 0 \\ -2\Gamma_0 e \gamma \frac{1}{2 + \gamma} & eV < 0 \end{cases}
$$
 (1)

for the case of positive and negative bias, respectively. In this expression, the current depends on the gate voltage through the factor γ , and can be directly compared to the experimental data. Figure $4(a)$ shows that the negative-bias current calculated from Eq. [\(1](#page-1-2)) is in excellent agreement with the measured negative-bias current as a function of gate voltage. The positive-bias current is analyzed in Fig. [4\(b\)](#page-2-3) for three different values of polarization *p*: 0.0, 0.3, and 0.4. We find the calculated current for the $p = 0.0$ case does not fit the data, while there is a good agreement when the current is calculated with a *p* value between 0.3

FIG. 2 (color online). (a) Differential conductance, $G =$ dI/dV , for varying gate and bias voltages with regular coulomb diamonds. Black represents zero conductance and yellow indicates a maximum conductance of 0.06 e^2/h . Dashed lines indicate the bias voltage of 5 mV. (b) Conductance as a function of gate voltage at 5 mV. Open blue circles indicate negative bias while full green triangles represent positive bias. Positive-bias data have been shifted in the gate voltage by $\Delta V_G =$ 10 mV/ α = 0.34 V to allow direct comparison between corresponding conductance peaks. Theoretical fits (represented by solid lines) come from the Breit-Wigner model with locations of conductance peaks as a fitting parameter.

FIG. 3. (a) Normal-lead conductance for positive (full diamonds) and negative (open diamonds) bias. Positive-bias data have been scaled by a factor of 1.15 so that $G_{N+} = G_{N-}$. (b) Ferromagnetic-lead conductance for positive (full circles) and negative (open circles) bias. Positive-bias data (G_{FM+}) have been scaled by the same factor as G_{N+} .

and 0.4. This result agrees with the expected thermodynamic polarization value for cobalt, $p = 0.4$ [\[29\]](#page-3-23).

We further analyze the deviation of the positive-bias current from the $p = 0.0$ curve by defining the current difference, ΔI , as $\Delta I = (I_0 - I_+)/(I_0)$, where I_0 is the calculated current for the $p = 0.0$ case and I_+ is the measured current for positive bias. This current difference depends strongly on the gate voltage, with peaks ranging from 8% to 17%, as shown in Fig. [4\(c\).](#page-2-3) It should be emphasized that ΔI shown in Fig. $4(c)$ describes only the deviation from the $p = 0.0$ line, and is only observed in the case of positive bias.

It is important to note that the current difference ΔI peaks for the values of gate voltage at which the normal lead is resonant with the single energy level on the dot. In this situation, the conductance is dominated by the nonresonant tunnel junction between the ferromagnetic electrode and the quantum dot. The tunneling rates for the majority and minority spins are different $(\Gamma_{FM}^{\dagger} \neq \Gamma_{FM}^{\dagger})$ for this junction, meaning that there are fewer available states for the minority spins in the ferromagnetic electrode. As a result, the minority spins will spend more time on the dot, leading to spin accumulation [\[15\]](#page-3-9). This leads to a decreased current due to electrons with minority spins, resulting in a decreased total conductance through the device for positive bias. This spin-diode effect depends on the fact that there is a single nondegenerate level available on the dot in the bias window. As described theoretically in detail in Refs. [[11](#page-3-21),[15](#page-3-9)], our results imply that the current through the device is spin-polarized for specific values of gate and bias voltages. When the gate and bias voltage are tuned away from this regime, the current rectification is not observed, and the current is not spin polarized. In particular, when the bias is increased to allow transport through another energy level on the dot, the spin-diode effect is diminished or destroyed by spin flips.

In summary, we have observed a clear asymmetry in the current through a quantum dot with one ferromagnetic and one normal-metal lead as a function of bias direction. Our

FIG. 4. (a) Negative-bias current (absolute) as a function of gate voltage. Open black squares represent measured current. Solid line is a fit obtained from Eq. [\(1\)](#page-1-2) for $eV < 0$ case. (b) Positive-bias current as a function of gate voltage. Open black squares represent measured current. Theoretical fits to the data use $eV < 0$ case of Eq. ([1\)](#page-1-2) with $p = 0.0$ (dotted red), 0.3 (dashed blue), and 0.4 (solid green). Conductance ratio, γ , in Eq. ([1](#page-1-2)) is taken from the negative-bias data in Fig. [3.](#page-2-4) (c) ΔI as a function of gate voltage for a fixed bias voltage (5 mV) . ΔI calculated as difference from theoretical $p = 0.0$ fit in Fig. [4\(b\).](#page-2-3)

analysis shows that this rectification occurs because of the asymmetric tunneling rates between the ferromagnetic lead and the dot for the majority and minority spins. The observed asymmetry implies the existence of spinpolarized currents, which are fully tunable by a proper choice of gate and bias voltages. By increasing the polarization of the ferromagnet, the diode effect could be increased up to a ΔI value of 100% [[30](#page-3-24)]. Such carbonnanotube spin diodes could easily be positioned using ac dielectrophoresis [[24](#page-3-17)], and could be operated at room temperature by reducing their size [\[31\]](#page-3-25). Additionally, with the proper choice of the material for the nonmagnetic electrode [\[32\]](#page-3-26), this device could be invaluable for achieving controllable spin injection.

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^[1] I. Zutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

^[2] X. Lou, C. Adelmann, S. A. Crooker, E. S. Garlid, J. Zhang, K. S. M. Reddy, S. D. Flexner, C. J. Palmstrom, and P. A. Crowell, Nature Phys. **3**, 197 (2007).

- [3] I. Appelbaum, B. Huang, and D.J. Monsma, Nature (London) **447**, 295 (2007).
- [4] M. Johnson and R. H. Silsbee, Phys. Rev. Lett. **55**, 1790 (1985).
- [5] D. J. Monsma, R. Vlutters, and J. C. Lodder, Science **281**, 407 (1998).
- [6] R. Fiederling, M. Keim, G. Reuscher, W. Ossau, G. Schmidt, A. Waag, and L. W. Molenkamp, Nature (London) **402**, 787 (1999).
- [7] Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, H. Ohno, and D. D. Awschalom, Nature (London) **402**, 790 (1999).
- [8] P. Recher, E. V. Sukhorukov, and D. Loss, Phys. Rev. Lett. **85**, 1962 (2000).
- [9] E. Cota, R. Aguado, C. E. Creffield, and G. Platero, Nanotechnology **14**, 152 (2003).
- [10] R. Świrkowicz, J. Barnaś, M. Wilczyński, W. Rudziński, and V. K. Dugaev, J. Magn. Magn. Mater. **272**, 1959 (2004).
- [11] W. Rudziński and J. Barnas, Phys. Rev. B 64, 085318 (2001).
- [12] B. R. Bulka, Phys. Rev. B **62**, 1186 (2000).
- [13] M. Chshiev, D. Stoeffler, A. Vedyayev, and K. Ounadjela, Europhys. Lett. **58**, 257 (2002).
- [14] W. Kuo and C. D. Chen, Phys. Rev. B **65**, 104427 (2002).
- [15] F. M. Souza, J. C. Egues, and A. P. Jauho, Phys. Rev. B **75**, 165303 (2007).
- [16] H. Dalgliesh and G. Kirczenow, Phys. Rev. B **73**, 235436 (2006).
- [17] A. A. Shokri, M. Mardaani, and K. Esfarjani, Physica E (Amsterdam) **27**, 325 (2005).
- [18] J. Wang, K. S. Chan, and D. Y. Ying, Phys. Rev. B **72**, 115311 (2005).
- [19] S. Ijima, Nature (London) **354**, 56(1991).
- [20] R. Saito, G. Dresselhaus, and M. S. Dresselhaus, *Physical Properties of Carbon Nanotubes* (Imperial College, London, 1998).
- [21] N. Tombros, S. J. van der Molen, and B. J. van Wees, Phys. Rev. B **73**, 233403 (2006).
- [22] M. Bockrath, D. H. Cobden, P. L. McEuen, N. G. Chopra, A. Zettl, A. Thess, and R. E. Smalley, Science **275**, 1922 (1997).
- [23] J. Kong, H. T. Soh, A. M. Cassell, C. F. Quate, and H. Dai, Nature (London) **395**, 878 (1998).
- [24] K. Yamamoto, S. Akita, and Y. Nakayama, J. Phys. D **31**, L34 (1998).
- [25] C. W. J. Beenakker, Phys. Rev. B **44**, 1646 (1991).
- [26] H. Grabert and M. H. Devoret, *Single Charge Tunneling* (Plenum, New York, 1992).
- [27] W. Liang, M. Bockrath, and H. Park, Phys. Rev. Lett. **88**, 126801 (2002).
- [28] L. I. Glazman and K. A. Matveev, JETP Lett. **48**, 445 (1988).
- [29] D. J. Monsma and S. S. P. Parkin, Appl. Phys. Lett. **77**, 720 (2000).
- [30] K. P. Kamper, W. Schmitt, G. Guntherodt, R. J. Gambino, and R. Ruf, Phys. Rev. Lett. **59**, 2788 (1987).
- [31] H.W.C. Postma, T. Teepen, Z. Yao, M. Grifoni, and C. Dekker, Science **293**, 76 (2001).
- [32] B.-C. Min, K. Motohashi, C. Lodder, and R. Jansen, Nat. Mater. **5**, 817 (2006).