Electrical Read-Out of a Single Spin Using an Exchange-Coupled Quantum Dot

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ABSTRACT: We present an original way of continuously reading-out the state of a single electronic spin. Our detection scheme is based on an exchange interaction between the electronic spin and a nearby read-out quantum dot. The coupling between the two systems results in a spin-dependent conductance through the read-out dot and establishes an all electrical and nondestructive single spin detection. With conductance variations up to 4% and read-out fidelities greater than 99.5%, this method represents an alternative to systems for which spin-to-charge conversion cannot be implemented. Using a semiclassical approach, we present an asymmetric exchange coupling model in good agreement with our experimental results.

KEYWORDS: quantum dot, single-molecule magnets, nanospintronic, single-molecule transistor, single-spin detection

Over the last 10 years, advances in nanofabrication and measurement technologies allowed for the read-out and manipulation of single electronic and nuclear spins. Besides the opportunity of testing our understanding of quantum mechanics, these progresses are at the heart of recent developments toward potential applications in the field of nanospintronics, molecular spintronics, and quantum information processing. Among different concepts, systems integrating all electrical spin detection benefit most from achievements of the microelectronic industry, but so far, they relied on the spin to charge conversion, which required emptying the electron into a nearby reservoir. However, in devices where the energy of the spin system is much more negative than the Fermi energy of the leads, a different detection technique is mandatory. Here we present a general detection scheme based on the exchange coupling between a single electronic spin coupled to a read-out quantum dot. Depending on the strength of the exchange coupling, this detection scheme allows an all electrical and nondestructive read-out of a single electronic spin, as the conductance of the read-out quantum dot is spin dependent.

To implement this general detection scheme, we made use of the electromigration technique to fabricate a single TbPc2 molecule spin-transistor (Figure 1a), that can be considered as two coupled quantum systems:

(i) First, an electronic spin carried by the terbium Tb3+ ion. Its total spin \( S = 3 \) and total orbital momentum \( L = 3 \) originate from its \([\text{Xe}]^{4f^8}\) electronic configuration. A strong spin–orbit coupling leads to a total angular magnetic moment \( J = 6 \) of the electronic spin. Moreover, the two phthalocyanines (Pc) generate a ligand field, leading an electron-ground state doublet \( |\uparrow\rangle \) and \( |\downarrow\rangle \). It results in an electronic-spin exhibiting a strong uniaxial anisotropy axis perpendicular to the plane defined by the phthalocyanines, as depicted in Figure 1a. At finite magnetic fields, the degeneracy of the doublet is lifted, the electronic spin can undergo a reversal by emitting a phonon via a direct relaxation process.

(ii) Second, the Pc ligands create a read-out quantum dot. The TbPc2 has a spin \( S = 1/2 \) delocalized over the two Pc ligands which is close in energy to the Tb-4f states. Thus, the delocalized \( \pi \)-electron system results in a quantum dot in the vicinity of the electronic spin carried by the Tb3+ ion. This read-out quantum dot is tunnel-coupled to source and drain terminals to perform transport measurements. As demonstrated in the following, a strong exchange interaction in between the delocalized \( \pi \)-electron and the Tb3+ electronic spin can be
observed, resulting in an electrical read-out of a single electronic spin without affecting its magnetic properties.

RESULTS

Now, we present the characterization of our single-molecule spin transistor measuring the differential conductance as a function of the source drain voltage \( V_{ds} \) and the gate voltage \( V_g \) at an electronic temperature around 80 mK to obtain the stability diagram presented in Figure 1b. Regions colored in red and blue exhibited respectively high and low differential conductance values. Note that this sample is the same as previously studied,\(^4,22\) the difference in conductance originating from a slight change of the molecules tunnel-coupling to the metallic leads due to aging of the device. From the general characteristics of the Coulomb diamond in Figure 1b, we obtained a conversion factor \( \alpha = \Delta V_{ds}/\Delta V_g \approx 1/8 \), resulting in a low estimation of the charging energy \( E_C \approx 100\text{meV} \) of the quantum dot under investigation. First, this large value agrees with the idea depicted in Figure 1a that the single TbPc\(_2\) molecular magnet can be the quantum dot under investigation. Moreover, it was proved by Zhu et al.\(^24\) that electrons added to the TbPc\(_2\) preferentially go to the Pc ligands up to the fifth reduction and second oxidation. Thus, as observed in our previous works on two different samples\(^4,11,22\) the charge state as well as the magnetic properties of the Tb\(^{3+}\) ion are conserved. On these complementary accounts, the read-out quantum dot is most likely created by the Pc ligands.

The stability diagram presented in Figure 1b exhibits a zero bias anomaly on the right side of the charge degeneracy point, which is associated with the usual spin \( S = 1/2 \) Kondo effect observed in 2D electron gas quantum dots\(^25,26\) and single molecule transistors.\(^28,29\) The Kondo temperature \( T_K \) was determined by measuring the differential conductance at \( V_{ds} = 0\text{ V} \) as a function of the temperature \( T \) for a fixed gate voltage

\[
V_g = -0.69\text{ V} \quad \text{(Figure 2a)}
\]

By fitting the results to the empirical formula:\(^25\)

\[
g(T) = \frac{T^2}{T_K^2}(2^{1/8} - 1 + 1) + g_c
\]

where \( g_0 \) is the maximum conductance, \( s = 0.22 \), and \( g_c \) is the fixed background conductance; we obtained a Kondo temperature \( T_K = 5.3 \pm 0.05\text{ K} \).
To determine the configuration and value of the exchange coupling between the electronic spin and the read-out quantum dot carried by the Tb\(^{3+}\) ion, we investigated the evolution of the Kondo peak depending on the applied bias voltage \(V_d\) and the external magnetic field \(B\). By increasing \(B\), the zero bias anomaly splits linearly, the value of the slope being 124 \(\mu\)V/T, as presented in Figure 2b. This slope is a direct measurement of the g-factor = 2.15 ± 0.1, which is consistent with the usual spin \(S = 1/2\) Kondo effect.

For a standard spin \(S = 1/2\) Kondo effect, an extrapolation of this linear splitting should give an intersection at a positive value of magnetic field \(B_\perp\) where \(B_\perp\) is the critical magnetic field, and is a direct measurement of the Kondo temperature \(T_K\) via \(\Delta g_\mu_B B_\perp = k_B T_K\). From the measurement presented in Figure 2b, we clearly observe a different behavior. An extrapolation results in a negative critical magnetic field \(B_\perp \approx -880\) mT. To understand this behavior, we use the analogy to the underscreened spin \(S = 1\) Kondo effect\(^{27,31}\) for which the Kondo resonance separates into Zeeman states for \(2g_\mu_B B_\perp \ll k_B T_K\). This effect originates from the ferromagnetic coupling of the remaining unscreened spin \(S = 1/2\), reducing the strength of the antiferromagnetic coupling between the screened spin \(S = 1/2\) and the electrons of the leads. Acting as an additional effective magnetic field, this ferromagnetic coupling lowers the value of \(B_\perp\).

In our single molecular magnet-based transistor, the Tb\(^{3+}\) ion carries an electronic spin with a magnetic moment equal to 9 \(\mu_B\). The negative value of \(B_\perp\) originates from the ferromagnetic coupling between the read-out quantum dot and this electronic spin. Taking into account this coupling, the relation between the critical field \(B_\perp\) and the Kondo temperature \(T_K\) can be modified to:

\[
2g_\mu_B B_\perp = k_B T_K + a \mu_B j_z S_z \tag{2}
\]

where \(a\) is the coupling constant, \(J_\perp\) and \(S_z\) are the \(z\) component of the electronic Tb\(^{3+}\) and read-out quantum dot spins, respectively. Using the Kondo temperature \(T_K = 5.3\) K obtained from eq 1 and the critical field extracted from the magnetic field dependence (Figure 2b), a coupling constant \(a = -3.91\) T is obtained. We emphasize that such a high value cannot be explained by a purely dipolar interaction due to the terbium magnetic moment. Indeed, the relative distance between the phthalocyanine read-out quantum dot and the Tb\(^{3+}\) ion is about 0.5 nm, giving a dipolar interaction of the order of 0.1 T, which is more than 1 order of magnitude lower than the measured coupling constant. As an efficient exchange interaction requires an overlap of the wave functions between the electronic magnetic moment carried by the Tb\(^{3+}\) ion and the read-out quantum dot, this high coupling further validates the expected configuration for which the read-out quantum dot is the phthalocyanine. We present in the Supporting Information two other TbPc\(_2\)-based spin transistors for which the exchange coupling was measured.

We now present the measurements and the model to explain how the exchange coupling between the electronic spin state and the read-out quantum dot induces a spin dependence of the differential conductance. We first define \(B_\parallel\) and \(B_\perp\) being the magnetic fields applied parallel and perpendicular to the easy axis of the molecule, respectively (Figure 1a). For \(V_d = 0\) and \(B_\perp = 0\), we recorded the differential conductance at the working point while sweeping \(B_\parallel\) (Figure 3a). By repeating this measurement, we obtained two distinct magneto-conductance signals, corresponding to the two electronic spin states \(\uparrow\) (red) and \(\downarrow\) (blue). The two measurements intersect at \(B_\parallel = 0\) and have a constant differential conductance difference for \(B_\parallel > \pm 100\) mT. To quantify the read-out fidelity of our device, we recorded the conductance values at \(B_\parallel = 100\) mT for 10000 measurements. Plotting the results into a histogram yielded two distinct Gaussian-like distributions as presented in Figure 3b. The read-out fidelity was determined to be higher than 99.5% by relating the overlap of the best fits to these two distributions.

To further characterize the signal originating from the electronic spin, we determined the conductance difference between the two different orientations of the electronic spin (\(\uparrow\)) as a function of \(B_\parallel\) and \(B_\perp\) (Figure 3c). Two different regions are clearly visible. In the red region, the conductance corresponding to spin \(\downarrow\) is lower than that for spin \(\uparrow\). In the blue region, we observe the inverse scenario. However, at a particular combination of \(B_\parallel\) and \(B_\perp\) the signal goes to zero, which is indicated by the white region. The dotted line in Figure 3c emphasizes the cross-section presented in Figure 3a.
\[ \mathcal{H} = \mathcal{H}_f + \mathcal{H}_s + \mathcal{H}_{\text{J}}, \]

\[ = \mathcal{H}_f + \mathcal{H}_{\text{read}} \]

\[ = \mu_0 B \cdot \mathbf{J} + \mu_0 B \cdot \mathbf{\bar{r}} \cdot \mathbf{s} + J \cdot \mathbf{\bar{r}} \cdot \mathbf{s} \]

where \( \mathbf{\bar{r}} \) and \( g_\alpha \) respectively are the g-factor of the read-out quantum dot and of the electronic spin, \( \mathbf{\bar{r}} \) is the exchange coupling, and \( \mu_0 \) is the Bohr magneton. In the experiment, the magnetic field \( B \) is applied along two directions, such that it can be defined in the \( x-y \) plane of the TbPc2 molecular magnet, resulting in \( B = (B_y, 0, B_z) \). Furthermore, \( J_x = \pm 6 e \) is considered as a classical vector confined on the easy axis of the TbPc2 molecular magnet. Because of the axial symmetry of the system, we consider it as invariant under a rotation in the \( x-y \) plan. The read-out dot Hamiltonian can be consequently defined in the \((L, ||)\) basis as

\[ \mathcal{H}_{\text{read}} = \mu_0 \left( \frac{B_1}{B_1} \frac{B_1}{B_1} \mathbf{\bar{r}}_s \left( s_{\|} \right) + \left( \frac{J_1}{J_1} \right) \mathbf{\bar{r}}_s \left( s_{\|} \right) \right) \]

(3)

According to the exchange tensor \( \mathbf{\bar{r}} \) and the g-factor in the \((L, ||)\) basis is

\[ \mathbf{\bar{r}} = \left[ \begin{array}{c} g_{\|} + \delta g_{\|} g_{\|} \\ \delta g_{\|} g_{\|} \end{array} \right] \]

(4)

where the notation “\( \delta \)” is used for the anisotropic contributions. Subsequently, taking \( s_{\perp} = \hbar \sigma_{\perp}/2 \) and \( s_{\|} = \hbar \sigma_{\|}/2 \), the Hamiltonian \( \mathcal{H}_{\text{read}} \) in the read-out dot electronic spin basis is given by

\[ \mathcal{H}_{\text{read}} = \frac{h \mu_0 g_{\|} B_1}{2} \left( 1 + \frac{\delta g_{\|}}{g_{\|}} \frac{B_2}{B_1} \right) \]

\[ + \frac{h \mu_0 a}{2} \left( 1 + \frac{\delta g_{\|}}{g_{\|}} \frac{B_1}{B_1} \right) \]

(5)

The eigenenergies of the read-out dot are

\[ E_{\pm 1/2} = \pm \left[ \epsilon_0 + 2\Delta g \mu_0 B g_{\|} J_z + 2\Delta J \left( g_{\|} + g_{\|}\right) \mu_0 B g_{\|} J_z \right]^{1/2} \]

(6)

where \( \epsilon_0 \) is function of \( J_z \), meaning that the states \( J_z = \pm 6 \) are degenerated for \( B_0 B_1 = -\Delta g (g_{\|} + g_{\|}) B_1 \). This results in a shift of the crossing point in \( B_y \) as the function of \( B_z \) observed in the measurement presented in Figure 3c, given by

\[ B_{y}^{\text{shift}} = \frac{g_{\|} + g_{\|}}{g_{\|}} \frac{\Delta J}{a} \]

(7)

To obtain an estimation of the off-diagonal term \( \Delta J \) as well as the anisotropy of \( g_{\|} \) we use the experimental values determined from the measurements presented in Figure 2 \( (a = -3.91 \, \text{T} \text{ and } g \approx 2.15) \), and extract the slope of \( B_{y}^{\text{shift}} = -0.6B_1 \) from the measurement presented in Figure 3c. We calculated and present in Figure 4 the different doublet \( \left( \frac{g_{\|}}{g_{\|}} \right) \) in accordance with the experimental measurements. An infinite number of doublets gives a perfect agreement with the experiment. Minimizing the anisotropy of the g-factor, we use \( \left( \frac{g_{\|}}{g_{\|}} \right) = 0, \frac{\Delta J}{a} = 0.6 \), we obtain the energy difference of the read-out quantum dot: \( \Delta E_{\text{read}} = E_{\text{read}} - E_{\text{read}} \), depending on the state \( | \uparrow \rangle \) or \( | \downarrow \rangle \) of the electronic spin, as a function of \( B_z \) and \( B_y \) (Figure 3d). The zero sensitivity region (in white in Figure 3c) as well as the qualitative agreement reinforce the model used to interpret the dependence of the magneto-conductance signal as a function of the electronic spin orientation.

### CONCLUSIONS

In summary, we report on the proposition, theoretical explanation, and experimental realization of an electrical read-out of a single electronic spin using an exchange coupled read-out quantum dot. This experimental realization has been demonstrated using the net magnetic moment of a single molecule, the read-out quantum dot being directly sensitive to the spin orientation resulting in signal amplitudes up to 4% and read-out fidelities of 99.5%. This detection scheme is fully consistent with any single molecule architecture for which the magnetic moment is carried by a single atom embedded by a ligand, as far as the charge state of the spin dot remains unchanged, as it is the case for all of the lanthanide double-decker family (Tb, Dy, Ho, etc.), and could also allow the detection of a single magnetic impurity in semiconductor quantum dots, or single spin coupled to a nanotube or
nanowire, leading to potential progress in nanospintronic and quantum information processing.

METHODS

The single TbPc₂ spin-based transistor was fabricated using an electromigration technique at very low temperature. Using standard optical and e-beam lithography techniques, a gold nanowire on an Au/HOO₃ gate fabricated through atomic-layer deposition was prepared. After a cleaning procedure of the nanowire was performed with the use of acetone, ethanol, isopropyl alcohol solution, and oxygen plasma, a dilute dichloromethane solution of the TbPc₂ was deposited onto the nanowire at room temperature. The device was then connected on its sample holder, and enclosed in a high-frequency, low-temperature filter. It consists of 24 superconducting wires coated with Eccosorb, and enclosed in a CuNi tube of 1.5 mm external diameter. This nanowire ramping the voltage (10 mV/s) at 50 mK, and measuring its electronic temperature of about 50 mK. A nanogap was created in the nanowire and enclosed in a CuNi tube of 1.5 mm external diameter. The device was then connected on its sample holder, and enclosed in a high-frequency, low-temperature filter. It consists of 24 superconducting wires coated with Eccosorb, and enclosed in a CuNi tube of 1.5 mm external diameter. This nanowire at room temperature. The device was then connected on its sample holder, and enclosed in a high-frequency, low-temperature filter. It consists of 24 superconducting wires coated with Eccosorb, and enclosed in a CuNi tube of 1.5 mm external diameter.

ASSOCIATED CONTENT

Supporting Information

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Notes

The authors declare no competing financial interest.

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