Electrically controlled quantum dot based spin current injector

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Abstract

We present a proposal for a fully electrically controllable quantum dot based spin current injector. The device consists of a quantum dot that is strongly coupled to a ferromagnetic electrode on one side and weakly coupled to a nonmagnetic electrode on the other side. The presence of ferromagnetic electrode results in an exchange field that splits the dot level. We show that this exchange-induced splitting can lead to almost full spin polarization of the current flowing through the device. Moreover, we also demonstrate that the sign of the polarization can be changed by the gate or the bias voltage within a switching time in the nanosecond range. Thus the proposed device can operate as an electrically controlled, fast switchable spin current source, which can be realized in various state-of-the-art nanostructures.

Spin injection is a central problem in the field of spintronics, and the improvement of its efficiency and its control enhances the performance of spin based devices. Most commonly ferromagnetic electrodes are used as sources of spin polarization, directly injecting spins from a ferromagnet into a connected device. However, the spin injection strongly depends on the ferromagnetic-normal interface: material issues and problems such as conductance mismatch (2, 3) make the realization of these boundaries technologically challenging. As an interface between the ferromagnet and the normal part tunnel barriers are widely used. The polarization of the injected spin current is then limited by the polarization of the tunneling electrons, which – for a typical ferromagnet – is in the range of 30-40%. Another drawback of this configuration is that the sign of
the current spin polarization can only be changed by rotating the magnetization of the ferromagnet, e.g. by applying an external magnetic field. Therefore, the spin polarization can be switched only relatively slowly, and in many cases the application of a local magnetic field is needed.

In this paper, we consider a device of different geometry, where the conventional tunnel barrier is replaced by a quantum dot (QD) coupled to a ferromagnetic (F) and a normal (N) lead (F/QD//N geometry), see Figure 1. We focus on the case, where the coupling to the ferromagnet is strong, while the coupling to the normal metal is relatively weak. Based on accurate numerical renormalization group calculations (NRG) we show that then the performance of the spin injection in such a quantum dot interface is greatly improved: First, the dot acts as a spin current amplifier, i.e., the spin polarization of the injected current highly exceeds that of the ferromagnetic electrode, and may even approach unity. Second, the polarization of the injected spin current becomes tunable by purely electric means, either by sweeping the gate voltage or simply by changing the applied bias voltage. Both ways enable extremely fast (∼1GHz) spin polarization switching. In this context, we discuss two geometries of our spin injection device. In the first, three-terminal device, we control the spin polarization by a gate voltage, while in the second, two-terminal set-up only the bias voltage is used for control. While the former geometry may be more relevant for semiconducting quantum dots, the latter is more suitable for molecular spintronics applications.

The principle of operation of the proposed spin polarization amplifier is based on the ferromagnetic exchange field induced level renormalization of the dot. When a quantum dot is coupled to external leads and the number of electrons on the dot is odd, strong electronic correlations can give rise to the spin formation and also to the Kondo effect. In case of ferromagnetic leads, quantum fluctuations renormalize the position of the dot levels differently for each spin direction due to the different spin-up and spin-down tunneling rates. This effective exchange field then splits the dot levels and suppresses the Kondo resonance. An important effect of the exchange field induced splitting is that the ground state as well as the local density of states of the dot become highly spin polarized. This is, in fact, the major ingredient of the efficient spin polarization amplification discussed here: due to the exchange splitting of the correlated state, the quantum dot will be able to offer spin current polarizations much higher than the ferromagnet itself. The second key ingredient is the asymmetry between the left and right contacts: the coupling to the ferromagnet should be larger than the coupling to the non-magnetic material to assure that only
highly spin polarized states carry the current in the transport window associated with the bias voltages. With more symmetrical couplings, one can still operate the device as an ultra-fast spin current injector, but polarizations will be reduced.

The proposed F/QD//N structure is quite generic, and our concept can be realized in various types of nanostructures using state-of-the-art nanotechnology. The basic ingredient, i.e. the ferromagnetic contact induced local exchange field [8, 11] has already been demonstrated experimentally in F/QD/F devices, where the QD was defined using fullerene molecules [10], self-assembled semiconductor nanocrystals [12], or carbon nanotubes [13, 14]. Very recently, the ferromagnetic contact induced spin splitting has been observed in F/QD/N devices, in InAs nanowire based QDs. [16]

We note that the problem of current spin polarization in quantum dots coupled to ferromagnetic and normal leads has already been studied both theoretically [17, 18, 19, 20, 21, 22] and experimentally [23, 24], however, mainly in the weak coupling regime. In this transport regime the effects related with the ferromagnetic-contact-induced exchange field are much suppressed and thereby the promising efficiency and functionality of the spin current injector and spin current amplifier discussed in this paper are missing.

Theoretical framework.— Our model consists of a quantum dot strongly coupled to the left (L) ferromagnetic lead and weakly coupled to the right (R) nonmagnetic lead (see Figure 1a). The strength of the tunnel couplings is described by the tunneling rates \( \Gamma_\alpha \) (\( \alpha = L, R \)), with \( \Gamma_L = (\Gamma_{L\uparrow} + \Gamma_{L\downarrow})/2 \). The spin polarization of the ferromagnet is taken into account by assuming different density of states for the spin-up (red) and spin-down (blue) subbands at the Fermi level, see Figure 1b. For the circuit in Figure 1c, the total electrostatic energy of the circuit is given by,

\[
E_{\text{tot}} = E(n) - eN_R V.
\]

Here the potentials of the left and right electrodes were explicitly set to \( V_L \to 0 \) and \( V_R \to -V \), and \( N_R \) denotes the numbers of particles on the right lead. The first term gives the energy of the quantum dot

\[
E(n) = E_C(n - n_g)^2 - \frac{1}{2} (C_R V^2 + C_g V_g^2), \tag{1}
\]

with \( n \) the number of particles on the dot, and \( C_\alpha \) (\( V_\alpha \)) the capacitances (voltages) of the left, right, and gate electrodes (\( \alpha = L, R, g \)), respectively. \( E_C = e^2/2C \) denotes the charging energy, with \( C = C_L + C_R + C_g \). The parameter \( n_g = (C_g V_g - C_R V)/|e| \) sets the number of electrons on
the dot, with $e < 0$ denoting the electron charge, and $V$ the voltage drop on the dot.

At the quantum level, the Hamiltonian of the system can be written as a sum of three terms, $H = H_L + H_R + T$. Here the left part $H_L$ describes the dot and the more strongly coupled ferromagnetic lead,

$$H_L = H_{0L} + E_C(n - n_g)^2 + t_L \sum_{k\sigma} (c_{Lk\sigma}^\dagger d_\sigma + d_\sigma^\dagger c_{Lk\sigma}) + \text{cst.},$$

and $H_R = H_{0R} - eN_RV$ describes the right electrode. In these equations $n$ and $N_\alpha$ are particle number operators, $n = \sum_\sigma d_\sigma^\dagger d_\sigma$, and $N_\alpha = \sum_{k\sigma} c_{\alpha k\sigma}^\dagger c_{\alpha k\sigma}$, with $d_\sigma^\dagger$ creating a spin-$\sigma$ electron on the dot and $c_{\alpha k\sigma}^\dagger$ being the creation operator of an electron of wave number $k$ and spin $\sigma$ in electrode $\alpha$. The Hamiltonian $H_{0\alpha} = \sum_{k\sigma} \varepsilon_{\alpha k\sigma} c_{\alpha k\sigma}^\dagger c_{\alpha k\sigma}$ describes noninteracting electrons in the leads, while the last term of $H_L$ describes tunneling between the electrons in the ferromagnetic lead and the dot with $t_L$ the corresponding tunneling amplitude. Finally, the tunneling between the left and right subsystem is described by

$$T = t_R \sum_{k\sigma} (c_{Rk\sigma}^\dagger d_\sigma + d_\sigma^\dagger c_{Rk\sigma}),$$

with $t_R$ the tunneling amplitude between the right lead and the dot.

Since the coupling of the dot is assumed to be much weaker to the right lead than to the left lead, we can perform a perturbative expansion in $T$. In leading order, we can express the current in spin channel $\sigma$ as

$$I_\sigma(V, V_g) = -\frac{|e|\Gamma_R}{\hbar} \int_{-\infty}^{\infty} d\omega A_\sigma(\omega, v_g - v) \left[f(\omega) - f(\omega - |e|V)\right],$$

where $v_g \equiv CV_g/|e|$ and $v = VC_R/|e|$ are the dimensionless gate and bias voltages, respectively (for the sign conventions, see Figure 1 and $A_\sigma(\omega, n_g)$ is the spin dependent spectral function of the quantum dot coupled to the ferromagnetic lead (F/QD), as described by Eq. (2). Notice that the second argument of $A$ determines the position of the dot level. Beside the dimensionless gate voltage, the level is also shifted by the bias voltage, which has a simple electrostatic reason (see Figure 1.c). This bias induced shift is important for the two terminal operation discussed below). The current is proportional to $\Gamma_R = 2\pi \rho_R t_R^2$, the tunneling rate to the right non-magnetic
electrode (assumed to have a constant density of states, $\rho_R$, and $f(\omega)$ denotes the Fermi function. Equation (4) implies that the spin polarization of the current, $P \equiv (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$, is determined by the difference of the spectral functions $A_\uparrow$ and $A_\downarrow$ in the energy window between the electrochemical potential of the left and right leads. As discussed in the following, the spectral functions depend strongly on the spin; due to the ferromagnetic lead induced exchange field, the two Hubbard peaks get spin polarized with opposite spin orientation (see Figure 1b). Large current polarizations can thus be achieved, if the left and right chemical potentials are positioned next to one of these peaks.

In order to calculate the spectral function of the F/QD system, we employ the numerical renormalization group (NRG) method. NRG is one of the most powerful tools to study transport through quantum dot structures, and it captures reliably the ferromagnetic exchange-field induced spin splitting of the dot levels. Note, that the accurate non-perturbative NRG approach is essential to describe the problem. Neglecting higher order correlations the spin amplification is lost, the output current polarization can not exceed the F lead polarization.

The NRG calculations were performed for the single impurity Anderson model, and the ferromagnet was modeled by flat subbands of different density of states for spin up and spin down electrons. We assumed parameters typical for quantum dot structures, and used $E_C/\Gamma_L = 10$, and a spin polarization of the ferromagnet $p = 0.4$. Having determined the spectral functions, we used Eq. (1) to compute the current. For the junction capacitances we assumed that $C_L/C_R = 2$, and $C_g/C_R = 0.1$, which are typical for semiconducting nanowire QDs.

The maximum value of the injected current is given by $I_0 = |e|\Gamma_R/\hbar$. Taking typical semiconducting QD parameters, $E_C = 5$ meV, $\Gamma_L = 0.5$ meV, and fulfilling the assumption of asymmetric couplings, i.e. $\Gamma_R = \Gamma_L/10$, yields the maximum current in the range of $I_0 \approx 50$ nA. However, if $E_C$ and $\Gamma_L$ are larger, as it is for e.g. carbon nanotube QDs, $I_0$ can be further enhanced. For molecule based QDs, where $E_C$ can be especially high ($E_C > 100$ meV), the maximum current can be even in the range of a few $\mu$A’s.

Spectral functions.– The results of our calculations for an unbiased dot ($V = 0$) are shown in Figure 2. Panel a(b) presents the colorscale plot of the normalized zero-temperature spin-up (spin-down) spectral function $A_\uparrow(\omega, n_g)$ ($A_\downarrow(\omega, n_g)$) with $\omega$ the energy measured from the Fermi level of the ferromagnet, and $n_g$ is the effective gate voltage. Upon increasing $n_g$, the dot’s
occupation changes from even to odd and then back to even. The change in charge occurs at
\( n_g \approx 1/2 \) and \( n_g \approx 3/2 \), where the neighboring charge states with odd and even electrons become
degenerate, and the Hubbard resonances reach the Fermi energy, \( \omega = 0 \). The region around \( n_g = 1 \)
is the Coulomb blockade region with an odd number of electrons (in our case \( \langle n \rangle \approx 1 \)). Here
additional lines associated with Kondo correlations appear at \( \omega \approx 0 \). In this regime, the dot
level is occupied by a single electron, and the conduction electrons try to screen it through the
Kondo effect. This would generate a resonance located normally at the Fermi level. \[6, 7\] However,
because of the ferromagnetic lead, an exchange field acts on the dot, and splits and suppresses the
Kondo resonance. \[5, 9\] This results in smaller resonances occurring at energies corresponding to
the magnitude of the exchange field. For the spin-up and spin-down orientations the resonances
shift in opposite direction, as clearly seen in the cross-sections of the colorscale plots, Figs. 2c–e.
The magnitude of the shift is equal to the effective ferromagnetic exchange field (\( B_{\text{exch}} \)) induced
Zeeman splitting, i.e. \( g\mu_B B_{\text{exch}} \). Importantly, due to correlation effects, this exchange field is not
constant, but monotonously changes with \( n_g \) (see Figure 2), and even reverses sign (see \( n_g \approx 1 \)),
as also demonstrated experimentally. \[13, 16\] This effect forms the basis of the proposed electrical
spin polarization control.

Besides the splitting of the Kondo resonance, the ferromagnetic exchange field has another,
even more dramatic consequence on the spectral functions: the Hubbard (resonance) peaks become
almost fully spin-polarized. For \( n_g < 1 \) (see Figure 2c,d) the lower/upper Hubbard peak is spin-up/spin-down polarized and when the exchange field changes sign (for \( n_g > 1 \)) the spin orientation
of the Hubbard peaks is also reversed, see Figure 2b. The proposed spin current injector is based
on this robust polarization of the Hubbard peaks. If the current flows e.g. in the energy window
shown by the gray stripe in Figure 2b, it is strongly polarized due to the much larger contribution
of the spin-down spectral density.

Three-terminal operation.– The current flowing through the F/QD//N device and its spin
polarization, \( P \), are calculated by plugging the NRG spectral functions into Eq. (1). The results
obtained are shown in Figs. 3a and b. The colorscale plot of the current (Panel a) shows the
expected Coulomb diamond behavior of the quantum dot. Focusing on the spin polarization (Panel
b), the quantum dot acts as a polarization amplifier: high current polarizations are achieved, which
could strongly exceed the polarization of the ferromagnetic lead (\( p = 40\% \)), and get close even
to full polarization. The spin polarization is strongly enhanced in an extended parameter region, especially close to the Coulomb peaks (i.e. around $v_g = 1/2, 3/2$) or, for larger bias voltages, outside the Coulomb diamond. As a spin current source, these regions are also preferable, since the high polarization is combined with high amplitude of the current. The sign of the spin polarization is parallel to the lead polarization in the red region, however, it can have opposite sign with similarly high amplitude as well (blue region). This change of polarization is driven by the sign change of the local exchange field, which induces in turn a polarization change of the occupied Hubbard peak, see Figure 2d-e.

Changing $v_g$ and thus the occupation by the gate voltage along the horizontal arrow in Figure 3b, e.g., the spin polarization of the current is reversed. This makes the F/QD//N device an efficient gate-controlled spin current source: the polarization is switchable by the gate electrode, which can be modulated really fast (over 10 GHz as shown by Nowack et al. [30]). The speed of the switch can also be limited by $\Gamma_L$, which is, however typically also in the GHz range or above, depending on the type of device.

As shown in Figure 3c, the switching of the spin polarization takes place for a wide range of applied bias voltages with relatively high up and down polarizations ($P \approx 90\%$). Figure 3d shows a proposed device geometry for the realization of the gate-controlled spin current source. A ferromagnetic lead couples strongly to a nanowire (or a carbon nanotube), a middle gate (MG) tunes the dot level, while a top gate (TG) defines the tunnel barrier with weak coupling, and the rest of the nanowire serves as a normal lead. Since the ferromagnetic lead induced local exchange field, its sign change, fabrication of side and top gates have all been demonstrated in carbon nanotube [13, 14] or nanowire [16] based structures, the realization of the proposal is within reach with state-of-the-art nanofabrication techniques. Besides the large amplification and the fast gate control, this geometry could solve the demanding materials issues of spin injection into carbon nanotubes or nanowires, i.e. the fabrication of proper tunnel barriers at the interface [11]: coupling a ferromagnetic lead strongly to these nanoobjects is much simpler, it induces a ferromagnetic proximity effect that gives rise to highly polarized spin current, which could be injected through the other weakly coupled barrier of the quantum dot.

Two-terminal operation. – The F/QD//N system can be also used as a two-terminal spin injector, where the presence of a gate electrode is not required. As it can be seen in Figure 3b, the value
of $v_g$, where the spin polarization reversal takes place depends on the bias voltage. The reason for this can be understood based on the simple circuit model shown in Figure 1c: the applied bias (V) also shifts the dot level with the value of $eV_d = -eV C_R/C$. As a consequence, the spin polarization can also be changed by just varying the bias voltage, e.g. along the vertical arrow shown in Figure 3b. We call this two terminal operation, since it does not require the change of $v_g$ i.e. the change of gate voltage, to reverse the spin polarization. As demonstrated in Figure 4, the spin polarization as function of the bias voltage reverses in a wide range of $v_g$. Moreover, polarization reversal with highly polarized spin-up and spin-down currents ($|P| > 0.6$) can be generated even with rather small bias voltages, $|V| < 0.4E_C/|e|$ (see the green curve in Figure 4a).

The two terminal operation allows the realization of the spin injector using molecular QD systems, such as e.g. modified buckyballs (see inset of Figure 4b) [31], where gates are not necessarily available. Due to the much larger energy scales of molecular quantum dots (charging energies of the order of tens - hundreds of meV), these devices could operate close to room temperature. Furthermore, molecular quantum dots also allow for the down-scaling of the spin injector, and support a much faster operation and a larger value of the injected current, too. In addition, molecules could also form monolayers, thus the output current of the spin injector could be enhanced significantly by the contribution of parallel molecular quantum dots.

Finally, let us comment on the presence of finite temperature, neglected so far. Since the injected current is almost entirely associated with the Hubbard peaks, the polarization amplification is effective as long as $T \ll E_C$. However, the polarization switch is smeared out by a finite temperature, and an effective exchange field $B_{\text{exch}} \gg T$ is required to polarize the dot spin and reach close to maximum polarizations. While for quantum dots this may imply below Kelvin device temperatures, for a molecular device these conditions can be relatively easily satisfied, and even a room temperature operation may be possible.

**Conclusions.**– In conclusion, we presented a proposal for an efficient spin injector, which is based on a quantum dot strongly coupled to a ferromagnetic lead and weakly coupled to a normal one. The proposed spin injector has several advantageous properties: (i) If the ferromagnet-dot coupling is substantially larger than the normal-dot coupling, then the output current gets almost fully polarized, since the dot strongly amplifies the spin polarization of the ferromagnetic lead. (ii) The polarization of the current can be reversed purely electrically by a small change of the gate
voltage. The spin switch is thus induced without modifying the polarization of the ferromagnetic lead, which allows an extremely fast polarization switching. (iii) The quantum dot based spin injector can also operate in a two-terminal configuration, where the current polarization is reversed by simply changing the applied bias voltage. This allows its implementation in molecular quantum dot systems as well. (iv) As a molecular electronics device, the proposed injector would allow the injection of relatively large polarized currents into other nanoscale objects, and could also be integrated in more complex nano-devices.

We emphasize again that the basic ingredients of the proposal have been already demonstrated experimentally in various nanoscale quantum dot systems, such as carbon nanotubes, semiconductor nanowires/dots or molecular quantum dots, thus the realization should be feasible with state-of-the-art experimental techniques.

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Figure 1: a) Device geometry: A quantum dot hosting odd number of electrons is placed at the interface of a ferromagnet (F) and normal contact (N). The ground state spin of the dot (down or up) determines the spin polarization of the current. b) Energy diagram of the device: The dot is strongly coupled to the ferromagnetic lead ($\Gamma_L$) and weakly coupled to the the normal lead ($\Gamma_R$). The ferromagnet-induced exchange field strongly polarizes the two resonance peaks of the dot. For the situation shown, the occupied (lower) Hubbard peak is spin-up polarized. The spin polarization of the current is generated by the polarization of the local density of states in the bias window. $V$ is the applied bias voltage and $E_C$ is the charging energy. c) Classical circuit diagram of the device: The quantum dot is capacitively coupled to the ferromagnetic and normal leads and to the gate electrode with $C_L$, $C_R$ and $C_g$, respectively. The gate voltage ($V_g$) allows to modify the level position. Due to the capacitances, the applied bias voltage ($V$) also shifts the dot level downwards, similar to the gate voltage, with a value of $eV_d = -eV C_R / C$, where $C = C_L + C_R + C_g$. 

\[ eV_d = -eV C_R / C \]
Figure 2: Spectral functions of a quantum dot strongly coupled to the ferromagnetic lead calculated by NRG. Panels a) and b) show the normalized zero-temperature spin-up ($A^\uparrow$) and spin-down ($A^\downarrow$) spectral functions as a function of energy ($\omega$) and effective gate voltage ($n_g$). Panels c), d), and e) show the cross-sections of the spectral function at the position of the dashed lines in a) and b) i.e. at $n_g = 0.7, 0.9, 1.1$, respectively. As can be seen, the lower (occupied) Hubbard peak is spin-up polarized for $n_g < 1$, while its polarization changes sign for $n_g > 1$. Crossing $n_g = 1$ either by gate or bias voltage induced level shift ($-eV_d$), the sign of the current polarization can be reversed. The gray area in Panel e) sketches the states contributing to the current at the bias voltage $V$. For the NRG calculation we used the parameters $p = 0.4$, $E_C = 0.1D$, $\Gamma_L = 0.01D$, and $D \equiv 1$ the half bandwidth.
Figure 3: Three terminal device operation: a) Total current $I = I_↑ + I_↓$ in units of $I_0 = |e| \Gamma_R / \hbar$ as a function of the dimensionless gate voltage, $v_g = C_g V_g / |e|$, and applied bias voltage $V$. $I/I_0$ shows the expected Coulomb diamond behavior. b) The spin polarization of the current for the same parameter range. As it is indicated by the white arrows the spin polarization can be reversed either by modifying the level position or changing the bias voltage. Panel c) shows the spin polarization characteristics as a function of dimensionless gate $v_g$ for different bias voltages. Panel d) shows schematic of the proposed realization of three terminal device: It consist of a nanowire (or a nanotube) strongly coupled to a ferromagnetic lead (F) with a top gate (TG) and a middle gate (MG). MG is used to change the level position of the dot, while TG defines the tunnel barrier with weak coupling. The calculations were performed for spectral functions shown in Figure 2 with $\Gamma_R / \Gamma_L = 0.1$. The capacitances were $C_L/C_R = 2$, and $C_g/C_R = 0.1$. 
Figure 4: Two terminal device operation: a) shows the spin polarization as a function of the bias voltage for different $v_g$. These are the cross-sections from Figure 3b. The spin polarization highly exceeds the polarization of the ferromagnetic lead and it reverses sign for positive bias voltages. Panel b) presents the proposed realization with asymmetrically coupled molecules.