Constructing Spin Interference Devices from Nanometric Rings

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The study of nanospintronic devices utilizing coherent transport through molecular scale multiply-connected geometries in the presence of moderate magnetic fields is presented. It is shown how two types of simple devices, spin filters and spin splitters (or Stern-Gerlach devices) may be constructed from molecular nanometric rings utilizing the Aharonov-Bohm effect. The current is calculated within a single electron approximation and within a many-body master equation approach where charging effects are accounted for in the Coulomb Blockade regime. We provide rules and tools to develop and analyze efficient spintronic devices based on nanometric interferometers.

I. INTRODUCTION

In recent years a great deal of attention has been devoted to the study of useful electronic devices utilizing the Aharonov-Bohm\(^1\) (AB) phase in multiply-connected geometries. In particular, there has been interest in spin-sensitive devices\(^2\) that are the single-electron analogue of semiconductor spintronics. Most of the research conducted in this direction has focused on mesoscopic systems, where the AB flux quantum matches weak magnetic fields, inter-electronic dependencies can be relatively negligible, and the Rashba\(^12\) and Dresselhaus\(^13\) spin-orbit coupling or inhomogeneous magnetic fields\(^\text{14-15}\) provide a large and controllable dependency of the electron Hamiltonian on directionally significant spin eigenvalues.\(^\text{16-18}\)

At the nanometric scale, it has been shown that spin-independent AB molecular interferometers may be possible at reasonable magnetic fields when the coupling of the device to the leads is small\(^\text{19-21}\). For such small AB interferometers there are striking differences in the properties of magnetic versus electric gauges, and the magnetic gate becomes advantageous over electrical gating.\(^\text{22-23}\) For example, the current in a multi-terminal molecular device can be tuned by changing the polarity of the magnetic field utilizing its symmetry breaking nature.\(^\text{24}\) Another example includes fundamental differences between magnetic and electric gates with respect to inelastic effects.\(^\text{25}\)

Previous studies of nanometric molecular AB interferometers have ignored the spin degree of freedom. In such molecular-scale systems, if the likes of atomic spin-orbit coupling and magnetic ions are not prevalent,\(^\text{26-28}\) the lack of a Rashba/Dresselhaus field and the difficulties in locally manipulating the external magnetic field at such scales leaves only the much smaller Zeeman term to differentiate between spins.\(^\text{29}\) In addition, the small size of such devices further complicates matters by introducing large charging effects.\(^\text{30}\) These should become especially noticeable when the coupling between the device and the leads is weak (as required for nanometric devices operating at reasonable magnetic fields), since electrons spend a longer time on the device. Despite these inconveniences, one may very well wonder whether it is possible to develop molecular AB interferometers as single-electron spin-devices such as a spin filter or splitter. This is the question that interests us here.

In this paper we develop the basic physical foundation to develop such devices. In Section II we describe the types of devices we wish to model and define their efficiency in terms of the spin-dependent current. This includes a two-terminal spin filter and a three-terminal spin splitter. In Section III Zeeman splitting is introduced into the two models developed by Hod et al.\(^\text{24-27}\) for independent spinless electrons - a simple continuum one-dimensional model and a more detailed tight-binding model. Within the simple continuum model we provide an exact solution for the spin-dependent conductivity for the two- and three-terminal devices. The role of the different model parameters is studied and comparison between the simple continuum model and the tight-binding results are made. In Section IV the tight-binding levels are used in a master equation calculation and to examine charging effects in the Coulomb blockade regime. From this basic analysis some interesting conclusions are drawn regarding the properties and limitations of nanospintronic AB devices, and a few potential avenues for further research are pointed out in Section V.

II. DEVICES

We will examine two types of elementary nanometric devices with and without charging effects: a spin filter and a spin splitter, as illustrated in Fig. 1. Such devices might be built from molecular rings such as the polycyclic aromatic hydrocarbons\(^\text{31-32}\) or from atomic corrals (with the advantage of controlled structure and electron density).\(^\text{33-34}\) Such corrals can be constructed atom by atom using scanning tunneling microscopy techniques.\(^\text{35}\) A more exotic example involves 3D nanostructures such as torus knots, where a periodicity of more than 2\(\pi\) can be achieved, producing a stronger AB effect than simple rings. In such molecular structures, control over the structure’s geometry is possible at the atomic level (un-
like the case of mesoscopic rings).

The filter does not really require something as complex as an AB ring for its realization. One only requires two single-spin levels which can, by manipulation of the magnetic field, be moved in and out of the conduction energy window, and any atom, quantum dot, or other discrete system can meet this requirement. This case is treated because it provides a simple example with only two terminals and yet may still be useful. The splitter is a molecular Stern-Gerlach device and is somewhat more interesting: having three terminals, it actually requires a device which, like an AB ring, has some sort of inherent asymmetry in either its construction or in the spinor wavefunction.

It is useful at this point to examine the energy scales of the problem. The Fermi energy is the most important parameter. For a half-filled conduction band the Fermi wavelength is of the order of four bond lengths \( \lambda_F = 4a \). This gives a Fermi energy of \( \epsilon_F = \frac{\hbar^2}{2m^* \lambda_F^2} \approx 10\text{eV} \) if we set the lowest level to zero, regardless of ring size and assuming atomic spacing of about \( a = 2\text{Å} \). To a good approximation, the molecular orbital energy (at zero magnetic field \( B = 0 \)) is given by an effective mass model

\[
\epsilon_m = \frac{\hbar^2 m^2}{2m^* R^2},
\]

where \( m = 0, \pm 1, \cdots \), \( R \) is the radius of the ring and \( m^* \) is the effective mass. This gives a value for the Fermi quantum number of \( m_F = \frac{2\hbar}{a} \). Electronic levels on the rings near the Fermi energy are separated by \( \Delta \epsilon = \frac{\hbar^2 \pi}{m^* R a} \) which reduces linearly with \( R \). For rings of the order of several nanometers in diameter the separation between levels near the Fermi energy is of the order of sub electron volts. This is also the order of magnitude of the charging energies \( \epsilon_c \). Other energy scales involve the magnetic field splitting which can be as large as the level spacing and the Zeeman splitting \( 2.89 \cdot 10^{-5}\text{eV} \) per Tesla, which is only about a third of the thermal energy at 1K.

The effectiveness of these devices can be judged not only by output polarization, but also by unitarity: a perfect device always sends the correct electrons into the correct lead. More precisely, the two devices that will be discussed are judged by the following quantities:

\[
e\uparrow\downarrow_{\text{filter}} \equiv \frac{j_1}{j_1 + j_2} \quad (1a)
\]

\[
e\uparrow\downarrow_{\text{filter}} \equiv \frac{j_1}{j_1 + j_2} \quad (1b)
\]

\[
e_{\text{splitter}} \equiv \frac{(j_1 - j_2)(j_1^2 - j_2^2)}{(j_1^2 + j_2^2)(j_1^2 + j_2^2)} \quad (1c)
\]

Here \( j_{1,2} \) is the up/down current for the two terminal device and \( j_{1,2} \) is the up/down current for channel 1 or 2 for the three terminal device. The only important properties of these quantities need to have for the purpose of this work is that they should be bounded from above by 1, and should reach 1 only in the case of perfect operation. For the two and three terminal devices both requirements are satisfied. We refer to them from now on as “efficiencies”.

The problem of building a perfect device can thus be mathematically restated as the problem of optimizing its efficiency to the desired value, usually unity, over the space of all controllable parameters. These include the magnetic field, the ring’s radius and structure, the gate and bias voltage (or the chemical potential) and the lead placement and coupling. Reasonable ranges for these parameters must be assumed: magnetic fields of more than a few Tesla may pose a technical limitation, as are large parameters which begin to dominate. The leads cannot be too close to one another to avoid direct tunneling and should be coupled strongly enough to make the environmental coupling unimportant. Even within these limits, the problem remains numerically formidable in all models. We will therefore also discuss a conceptually simpler way of designing perfect devices.

**III. SINGLE ELECTRON PICTURE**

In this section we describe the physical principles required to construct molecular spin filter and spin splitter devices in the limit where charging effects can be ignored and the complexity of the many-body physics can be reduced to a simplified one-electron picture.

**A. Basic Concepts**

To discuss the basic physical principles involved, let us first regard a simple analytical model for the transmission
through an AB ring.\textsuperscript{10,11,15,18,22} The physics of this model for molecular conductors was recently discussed by Hod et al. in the absence of Zeeman splitting and spin-orbit coupling.\textsuperscript{24,25,26,27} Consider a one dimensional AB interferometer as shown in Fig. 1. It consists of a conducting ring of radius $R$ coupled to two or three conducting wires placed in a perpendicular uniform magnetic field, $\mathbf{B}$. The device is described by the Pauli Hamiltonian:

$$H_s = \frac{\Pi^2}{2m^*} + V(\mathbf{r}) + g\mu_n\sigma \cdot \mathbf{B},$$

where $\Pi = P + \xi \mathbf{A}(\mathbf{r})$, $\mathbf{A}(\mathbf{r})$ is the vector potential ($\mathbf{B} = \nabla \times \mathbf{A}$), $V(\mathbf{r})$ is the electrostatic potential on the ring, and $m_e$ is the electron mass. The last term in Eq. (2) represents the coupling of the magnetic field to the spin angular momentum, where $\sigma$ are the Pauli matrices, $g$ is the gyromagnetic ratio (we take $g = 2$ for the spin), and $\mu_n = \frac{e\hbar}{2m_e}$ is the electron Bohr magneton. The lack of a Rashba field in molecular conductors allows us to safely ignore spin-orbit coupling effects. In the common case where the scalar potential $V(\mathbf{r})$ governing the system is periodic or nearly so, the effective mass approximation can be used and the Pauli Hamiltonian can be reduced to:

$$H_s = \frac{\Pi^2}{2m^*} + g\mu_n\sigma \cdot \mathbf{B} = H_{\text{ring}} + H_\sigma,$$

where $m^*$ is the effective mass of the electrons. Since the two additive terms in the Hamiltonian must commute (one depending only on the orbital part and the other on the spin part of the wavefunction), we can adopt the solution of Hod et al.\textsuperscript{24,25,26,27} for the transmission of electrons through the two prototype devices (spin filter and splitter). We adopt a transformation $\epsilon \rightarrow \epsilon - \epsilon_s$ which allows us to project spin effects onto a calculation where spin was previously neglected, noting that the spin dependent part of the Hamiltonian under a constant field must have the two eigenvalues $\epsilon_s = \pm g\mu_n B$ for spin up/down electrons. This transformation is not limited to the specific Hamiltonian described above: the only requirement is that $H_{\text{ring}}$ does not contain a spin dependency. Furthermore, even for inhomogeneous magnetic fields, where the spin-dependent term in the Hamiltonian does not commute with $H_{\text{ring}}$, a similar transformation in the limit of adiabatic spin dynamics can be made, where in addition to a shift in energy one has to introduce a shift in the magnetic flux.\textsuperscript{29}

Using the standard analytical approach of treating a 1D ring based on a scattering matrix formalism\textsuperscript{30,31} the transmission as a function of energy, previously calculated for spinless electrons\textsuperscript{32,33} need only be modified by the Zeeman energies for up (down) electrons, which affects only the kinetic phase angles $\phi_k$ in the expression:

$$\phi_k^{1\dagger} \equiv \phi_k(\epsilon_{1\dagger}) = \frac{\pi R}{\hbar}\sqrt{2m^*\epsilon_{1\dagger}} = \frac{\pi R}{\hbar}\sqrt{2m^*(\epsilon \pm g\mu_n B)}.$$  

The transmission itself is the solution of the linear scattering problems with the final results for the two-terminal device shown in the upper panel of Fig. 1 given by:

$$T_{1\dagger}(\epsilon) = \frac{A_{1\dagger}(1 + \cos 2\phi_m)}{R_{1\dagger}^{-1} + P_{1\dagger} \cos 2\phi_m + Q_{1\dagger} \cos 2\phi_m}.$$  

where $\phi_m = \pi \frac{\phi}{\phi_0}$ is the ratio between the magnetic flux $\phi = \pi R^2 B$ and the quantum flux $\phi_0 = \frac{2\pi h}{\epsilon}$, and we have defined:

$$A_{1\dagger} = 16\epsilon^2(1 - \cos 2\phi_k^{1\dagger})$$
$$P_{1\dagger} = 2(c - 1)^2(c + 1)^2 - 4(c^2 + 1)(c + 1)^2 \cos 2\phi_k^{1\dagger}$$
$$Q_{1\dagger} = (c + 1)^4$$
$$R_{1\dagger} = (c - 1)^4 + 4c^4 + 4 - 4(c^2 + 1)(c - 1)^2 \cos 2\phi_k^{1\dagger} + 8c^2 \cos 4\phi_k^{1\dagger},$$

with $\sqrt{\epsilon}$ the transmission amplitude into the junctions and $\epsilon = \sqrt{1 - 2\epsilon}$ the junction scattering amplitude.\textsuperscript{23}

A similar calculation can be made for the more cumbersome case of a three terminal device shown in the lower panel of Fig. 1. We focus on the case where all three junctions have identical scattering amplitudes. The transmittance for channels 1 and 2 is given by the ratio $T_{1\dagger,2}(\epsilon) = N_{1\dagger,2}(\epsilon)|D_{1\dagger,2}(\epsilon)|^{-1}$. The denominator of the transmittance probability for both output channels is given by

$$T_{1\dagger,2}(\epsilon) = \frac{N_{1\dagger,2}(\epsilon)|D_{1\dagger,2}(\epsilon)|^{-1}}{1 - |D_{1\dagger,2}(\epsilon)|^2}.$$
The numerator of the transmittance probability through output channel 1 is given by

\[
N_{11}^1(e) = -\frac{1}{2}e^2 \left\{ -4(1 + c^2)(2c - 1)^2 \cos(4\phi_k^{11}a) + (c + 1)^2 \cos(4\phi_k^{11}b) + 2(1 - c)^2 \cos(4\phi_k^{11}\gamma) + \\
4c \cos \left[ 4\phi_k^{11}(a + \gamma) \right] - (c - 1)^2 \cos \left[ 4\phi_k^{11}(a - \gamma) \right] - 2c(c + 1) \cos \left[ 2\phi_m - 2\phi_k^{11} \right] - 2(c + 1) \cos \left[ 2\phi_m + 2\phi_k^{11} \right] - \\
(c^2 - 1) \cos \left[ 2\phi_m + 2(1 - 2\alpha)\phi_k^{11} \right] + (c^2 - 1) \cos \left[ 2\phi_m - 2(1 - 2\alpha)\phi_k^{11} \right] + 2(c + 1) \cos \left[ 2\phi_m + 2(1 - 2\beta)\phi_k^{11} \right] + \\
2c(c + 1) \cos \left[ 2\phi_m - 2(1 - 2\beta)\phi_k^{11} \right] - (c^2 - 1) \cos \left[ 2\phi_m + 2(1 - 2\gamma)\phi_k^{11} \right] + (c^2 - 1) \cos \left[ 2\phi_m - 2(1 - 2\gamma)\phi_k^{11} \right] \right\}
\]
and the numerator of the transmittance probability through output channel 2 is given by

\[
N_{11}^2(\epsilon) = \frac{1}{2} \varepsilon^2 \left\{ -4(1 + \varepsilon^2) + (c + 1)^2 \cos(4\phi_k^i) + 2(c - 1)^2 \cos(4\phi_k^i) - 2(c - 1)^2 \cos(4\phi_k^i) \right\}
\]

where \(\epsilon = 2p - \alpha - \beta\) are the angles between all three leads as defined in Fig. 1.

The current is related to the transmittance through the Landauer formula. For the two terminal device the current is given by

\[
j_{11} = \frac{e}{\pi \hbar} \int_{-\infty}^{\infty} d\epsilon \left\{ f(\epsilon - \mu_I) - f(\epsilon - \mu_O) \right\} T_{11}(\epsilon) \tag{10}
\]

where \(\mu_I, \mu_O\) are the chemical potentials of the input/output channels and the Fermi function is \(f(\epsilon) = \frac{1}{1 + e^{(\epsilon - \mu)/T}}\). For the three terminal device the current is given by

\[
j_{11} = \frac{e}{\pi \hbar} \int_{-\infty}^{\infty} d\epsilon \left\{ f(\epsilon - \mu_I) - f(\epsilon - \mu_{O1}, \mu_{O2}) \right\} T_{11}^{12}(\epsilon), \tag{11}
\]

where \(\mu_{O1}, \mu_{O2}\) is the chemical potential for output channel 1 or 2, respectively. Conductance can be obtained from the current by taking the derivative with respect to the bias voltage.

\[\text{B. Spin Filters}\]

We now turn to discuss the application of the above results to the construction of a spin filter device. While for the case where the Zeeman effect was neglected, \(24,25,27\) it was always desirable to vary the gate voltage so as to shift the conduction peaks near zero magnetic field, in Zeeman spin devices this heuristic is complicated by the dependence of the splitting on the magnetic field strength. In practice, a compromise between realistically low magnetic fields and usable energy shifts restricts the sought after set of parameters. These include the kinetic phase angle \(\phi_k^i = \frac{\pi R}{\hbar} \sqrt{e \mu_i / m} \phi_m\) and the transmission amplitude into the junction \(\sqrt{\varepsilon}\) (cf., Eqs. \(5\) and \(6\)). Note that the kinetic phase angle depends on the magnetic phase angle \(\phi_m\). This is precisely where the aforementioned complication enters.

With the above convenient expressions one can calculate the spin-dependent and spin-independent conduction for different ring configurations and external parameters.

Typical results are shown in Fig. 2 for zero bias voltage \(\mu_I = \mu_O\) and for \(T = 0K\). As expected, the spin-independent conduction is periodic with a period that is equal to \(\phi_0\). Therefore, we plot only the first period, namely \(0 \leq \phi / \phi_0 \leq 1\). We find that within each period the spin-independent conduction has a symmetric structure around \(\phi / \phi_0 = 1/2\), characterized by a double peak.\(24,27\) This structure is caused by resonance transmission through the energy levels of the ring. The spin-dependent conduction follows closely the behavior of the total conduction. The up/down conduction peaks are separated by the Zeeman splittings which increase with increasing magnetic flux.

Similar to the case of spinless electrons changing the coupling strength between the leads and the ring, modeled here by \(\varepsilon\), changes the width of the peaks as shown in the left panels of Fig. 2. The effects of changing the kinetic phase angle are more involved than the case studied before.\(24\) In the case of spinless electrons, the kinetic phase angle depends only on the product \(R \sqrt{m/\hbar} \varepsilon\). Therefore, changing the energy of the conduction electron by the application of a gate voltage, changing the ring dimensions, and changing the effective mass of the conduction electron can be mapped onto a universal curve.\(24,27\)

In the present study there are three independent parameters that affect the kinetic phase angle in different ways. For example, the position of the conduction peaks can be shifted by adjusting the energy of the conduction electron by the application of a gate voltage. This is depicted in the lower left panel of Fig. 2. However, the change in the energy of the conduction electron also affects the splitting between the conduction peaks of the up/down spins as previously discussed. Altering the effective mass within the ring with the chemical potential held constant, for instance by a change of composition or inter-atomic distance in the ring atoms, modifies the kinetic phase and can thus increase or decrease the splitting as shown in the upper right panel of Fig. 2. Finally, as the ring radius is increased, the splitting for a set flux ratio decreases since this ratio then represents a smaller field as depicted in the lower right panel of Fig. 2. However, for the same reason, greater magnetic flux ratios become accessible for larger rings.

It is often more instructive to look at the results shown in the right panel of Fig. 2 where the familiar diamond-
magneto-conduction for a molecular device with related individual couplings between the leads and the ring ("bad contact"). In Fig. 3 we plot the magneto-conduction for a molecular device with relatively high effectiveness. We zoom on the region of the first magnetoconductance peak. The upper panel shows the results for a analytical model discussed above and the lower panel shows the results of an atomistic calculation for a molecular AB interferometer as illustrated in the upper panel of Fig. 4. As clearly can be seen, depending on the value of the magnetic gate, one can open output channel for spin up or down while at the same time close this channel for the other spin.

To calculate the magneto-conductance of a molecular AB interferometer we have used a simple tight-binding model where we assume a single electron in a spherical s-level for each site on the molecular ring. We add the proper magnetic terms to the tight-binding Hamiltonian of the system:

\[
H_s = H_{TB} + \mu_n \mathbf{\hat{L}} \cdot \mathbf{B} + \frac{1}{2} m_e \mu_n^2 B^2 R_1^2 + g \mu_n \mathbf{\sigma} \cdot \mathbf{B},
\]

where \( \mathbf{\hat{L}} \) is the angular momentum operator, \( \mathbf{B} \) is the magnetic field vector, and \( R_1 \) is the projection of \( \mathbf{R} \) onto the plane perpendicular to \( \mathbf{B} \). A gauge invariant basis is used to evaluate the tight-binding Hamiltonian matrix: 

\[
|1s\rangle_{\alpha} = |1s\rangle_e \exp \left(-\frac{\mu}{2} \mathbf{A} \cdot \mathbf{r} \right),
\]

where \( |1s\rangle_e \) is a 1s type orbital centered on site \( \alpha \), and \( \mathbf{A}_\alpha = -\frac{1}{2} (\mathbf{R}_\alpha \times \mathbf{B}_{\alpha}) \) is the vector potential evaluated at the position \( \mathbf{R}_\alpha \) of site \( \alpha \). We take the diagonal matrix elements of \( H_{TB} \) to be equal to zero (energy scale) and the off-diagonal elements are proportional to the overlap between the gauge invariant basis on the different electron sites, as described in more detail in Refs. 24 and 27.

The conductance is calculated using the Landauer formalism [23] which relates it to the scattering transmittance probability through the system. The transmittance is given by

\[
T(\varepsilon) = 4 \text{Tr} \left[ \hat{G}^{\dagger}(\varepsilon) \hat{\Gamma}_I(\varepsilon) \hat{G}(\varepsilon) \hat{\Gamma}_O(\varepsilon) \right].
\]

Here, \( \hat{G}(\varepsilon) = \left[ \mathbf{S} - H_s - i (\hat{\Gamma}_I + \hat{\Gamma}_O) \right]^{-1} \) is the retarded Green function, \( \mathbf{S} \) is the overlap matrix, and \( \hat{\Gamma}_I , \hat{\Gamma}_O \) are the imaginary parts of the self-energy (\( \Sigma \)) of the input/output channels. For the results presented in Fig. 3 we use both imaginary absorbing potential [23, 52] and an iterative semi-infinite bulk Green functions calculation scheme [25, 53, 54, 55] to calculate the self-energies of the leads.

Comparing the results of the simple analytical model to the results obtained from the tight-binding model indicates that the same physical picture emerges for the tight-binding approach. This is expected based on previous studies where the Zeeman effect was neglected [24, 25, 27]. The agreement between the two approaches indicates that the diffraction pattern is insensitive to the perturbations caused by an ionic potential and the results will not be invalidated by a more thorough (single-particle) treatment. The only free parameter used in the analytical theory is the scattering amplitude \( \sqrt{\varepsilon} \), which was adjusted to match the width of the conduction peaks. The effective mass entering the analytical model can be calculated directly from the tight-binding parameters. For a single s-level within tight-binding for a 1D crystal with inversion symmetry and a site distance of a one can show

\[
\text{FIG. 3: Plots of the conduction versus magnetic flux for the analytical model (upper panel) and for an atomistic calculation based on a tight-binding model (lower panel). The ring radius in both cases is } R = 5\text{nm}. \text{ Other parameters in the analytical model are } \varepsilon = \frac{1}{50}, \ m^* = 4m_e \text{ and } \lambda_F = 12\text{Å}. \text{Tight-binding parameters are defined in the text.}
\]

shaped conduction pattern within the energy/phase ratio plane behaves more simply and predictably, in order to gain a better intuitive understanding of the system. Here, the two ends of the white conduction lines represent up/down conduction where the chemical potential and magnetic field are at the middle of such lines. Varying \( \varepsilon \) still affects only the peak widths, modifying \( m^* \) changes the lengths of the conduction lines, changing \( m^* \) scales the diagram in the energy axis without changing the length of the conduction lines and altering the energy of the conduction electron scales the diagram along with the lines in the same axis. Thus, in order to construct an efficient spin filter one has to construct this diagram and control the position of the white conduction lines by the application of a proper magnetic field and gate voltages to achieve a desired behavior.

Good molecular spin filters can be made when two conduction peaks with different spins are sufficiently separated in energy and magnetic field to allow only electrons of one spin to traverse the ring. This is best achieved for molecular devices with a large effective mass for the conduction electron and small couplings between the leads and the ring ("bad contact"). In Fig. 3 we plot the magneto-conduction for a molecular device with relatively high effectiveness. We zoom on the region of the first magnetoconductance peak. The upper panel shows the results for a analytical model discussed above and the lower panel shows the results of an atomistic calculation for a molecular AB interferometer as illustrated in the upper panel of Fig. 4. As clearly can be seen, depending on the value of the magnetic gate, one can open output channel for spin up or down while at the same time close this channel for the other spin.

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\]

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that the dispersion relation can be approximated by\(^{(10)}\)
\[
\varepsilon_k \simeq E_0 + a^2 \gamma(a) k^2.
\]
This can be compared directly with a nearly free electron of mass \(m^*\) and energy \(\varepsilon_k = E_0 + \frac{k^2}{2m^*}\), allowing us to identify:
\[
m^* = \frac{\hbar^2}{2\gamma(a) a^2}.
\]
Here \(\gamma(a)\) is the resonance integral, i.e., the off-diagonal nearest neighbor matrix element of the tight-binding Hamiltonian.

C. Spin Splitter

The physics of the three-terminal device shown in the lower panel of Fig. 1 was recently discussed for the case where the Zeeman splitting was ignored\(^{(26)}\). Hod et al. showed how cyclic molecular rings can be used as parallel magnetoresistance logic gates (in contrast to the switching devices based on two-terminal rings)\(^{(22)}\). The basic idea was to couple the cyclic molecular system to three leads creating a three terminal device and to apply an external magnetic field. By carefully selecting a narrow resonance through which conductance occurred they showed that such a setup can be used to simultaneously switch one channel “on” and at the same time switch the other channel “off”. This was achieved by carefully adjusting the phase of the conducting electron with diminishing amplitude on one exit channel and a large amplitude at the other channel. A proper combination of a gate potential and output channels is optimized to achieve a desired efficiency such that a mirror symmetry around the magnetic field is always available (which appears to be the case), then using the simple analytical model of the ring this happens when:
\[
2g\mu_B B = \frac{\hbar^2}{2m^* R^2} [(m + 2)^2 - (m - 2)^2]
\]
Or, rewriting \(B\) as \(\frac{2a}{\phi_0}\) and \(\mu_B\) as \(\frac{e\hbar}{2m_e}\), we find that the condition where the Zeeman separation is equal to the separation between two ring energy levels is
\[
\frac{\phi}{\phi_0} = \frac{2m_e}{m^* g m_F} = \frac{2m_e \pi R}{gm^* a},
\]
where, as before, \(a\) is the separation between the sites on the ring. The condition given by Eq. 16 is a necessary but not sufficient condition for a spin splitting device. When the condition is met, the angles between the input and output channels are optimized to achieve a desired efficiency such that a mirror symmetry around the magnetic field given by Eq. 16 is achieved.

Similar sets of configurations with odd or half-integral flux ratio can be found in much the same way, due to the AB splitting. Clearly for rings with nanometer scale dimensions, where a flux ratio of one corresponds to thousands of Tesla, this is not a viable option unless \(m^* \gg m_e\). However, at tens of nanometers many configurations are possible at fields of a few Tesla. The main problematic issue that remains is the fact that in order to conduct through very low levels on the ring in this simple model, the Fermi energy must be lowered very significantly. If we assume that the Fermi level is of the order of \(\frac{\varepsilon_R}{e}\), one can show that in this simple model the magnetic field required to build this device at the Fermi energy becomes reasonable only when \(R\) is of the order of micrometers. However, if one manages to increase the
FIG. 4: Transmittance through a three-terminal ring device as a function of the flux ratio (magnetic field) and the kinetic energy of the conducting electrons. Left and right panels correspond to output channels 1 and 2, respectively. The ring has a radius of $R = 20\,\text{nm}$, which corresponds to an AB period of about 3Teslas. Upper panels: $m^* = 125\,m_e$, zero bias, and the two endpoints of the white line represent electrons of the two spins. Lower panels: $m^* = 10\,m_e$, small bias of 6meV, and here, spin-independent conduction occurs all along the purple line, while along the white (black) lines, still of length $g\mu_B$, only up (down) electrons are transmitted, as their conduction windows shift in energy according to their spins. Rainbow color scheme used (red indicates high transmittance of $\approx 1$).

spin g-factor\textsuperscript{46,47} or the effective mass in the device $m^*$ (see Eq. (14)), a nanometric device would be feasible at mere Teslas. This may hint that here as well a realistic device is a matter of the choice of materials - one would in principle need to custom-design a system in which the density of states is high enough at low kinetic energies that only levels of such low energies are occupied in the ground state, or where conduction sites are far apart and weakly connected\textsuperscript{46,47}.

The problems just mentioned are no longer present if one considers biased conduction (we assume the voltage falls symmetrically across the junctions to avoid complication) as shown in the lower panels of Fig. 4. The application of a finite bias voltage allows realistic configurations with the same structure at various effective masses. The example shown in the lower panels of Fig. 4 correspond to $m^* = 10m_e$ and a bias voltage of 6meV. The conduction window can be tuned to contain two quite distant levels. If levels with the transmissive properties of the ones previously discussed are selected, the Zeeman
FIG. 5: Plots of the spinless current versus magnetic field and gate voltage in the tight-binding approximation for the two exit channels of a 20nm 3-terminal ring device (left and middle panels). The spin dependent current is represented by the value of the current at the ends of the black lines. Right panel show the corresponding current effectiveness. In the right panel, color scale adjusted for maximum detail, such that the red areas represent a maximum effectiveness of approximately 0.6 (rainbow color scheme used). The best polarization achieved here is $\approx 90\%$ for one channel and $\approx 75\%$ for the other.

energy need not span the space between them. This can be done by the bias voltage and the Zeeman term must be no more than the level broadening (or a few $k_B T$ if this is more) to ensure conduction of only one spin per level. However, the conduction of electrons in the entire window of bias voltage should vanish in order to make efficient spin-splitter devices, since the biased current is the integral of the transmission over this energy range.

In an extension of the graphical method shown, the biased conduction window could be represented by a line of length $eV_B$ (purple line), where $V_B$ is the bias voltage. All “spinless” levels on this line conduct. Centered on the line's ends are two parallel lines of length $2g \mu_B B$ (black/white), where only one spin level is shifted into the window - therefore, at these ends only one spin type takes a part in conduction, while on the rest of the lines both do. Now, if this line is placed like the white lines in the upper panels of Fig. 4, with the levels at the ends in the one spin zones, the desired result is achieved as long as the conduction is zero for all other energies covered by the bias voltage. This can, however, be done at much higher energies and lower fields than those shown in the upper panels, since the level spacing spanned by the bias voltage can easily be orders of magnitude greater than $2g \mu_B B$.

In order to make a stronger argument that the spin-splitting configurations are a physical phenomenon rather than an idiosyncrasy of the simple one-dimensional analytical model used here, one might proceed by reproducing them in a more detailed numerical model. While remaining in the independent electron picture, an obvious and flexible choice is a ring of atoms represented by a tight-binding Hamiltonian, for which conductance can be calculated with the methods described above. The wealth of parameters makes it problematic to fine-tune exactly corresponding arrangements between the two models, and yet it is quite a simple matter to construct in one model a ring and lead configuration which worked well as a splitter in the other, and to try and see how well it works at some choice of parameters which should have analogous physical meaning. This should be more similar to what might be done in an actual experiment.

Since in the analytical model of a 20nm ring we have already located, as shown in Fig. 4, a good splitter configuration near 10 Tesla, the same configuration (with some effective mass) would be a convincing place to look for a splitter using the tight-binding model. We focus on the more realistic low effective mass regime where a bias voltage is needed and on a finite temperature of 1K, which should still leave the system well within the quantum regime. In Fig. 5 we plot the current as a function of the experimentally available parameters - the gate voltage and the magnetic field strength. The familiar asymmetric structure from the previous plots is blurred, but still readily recognizable in the two left panels, while the right panel shows the high effectiveness which can be reached when the magnetic field, the gate voltage and the bias voltage are all appropriately tuned. The location of the effectiveness peak could be easily predicted by the transmission line method previously discussed. Obviously, it would be practically impossible to stumble upon such a fortunate combination of conditions by accident, and any experiment must search for them under the explicit guidance of a model such as the analytical model suggested above.
IV. COULOMB BLOCKADE CALCULATIONS

A. Charging Energy

So far, we have ignored the energy it takes to inject multiple charges into the small region of the ring. This is of some concern since, as will be discussed below, the charging energy can be very significant here and especially when ring-leads coupling is weak, charging effects can play a major role. To stress this point, consider a device (ring) that is charged with an electron of spin up. The energy it takes to bring another electron with a spin down can be significantly different than in the single electron picture. Can this additional degree of freedom provide means to construct more efficient molecular spin filter and spin splitter devices, or will it prevent their realization?

In general, under our set of assumptions one can expect that when charging effects are neglected, states on the ring will be grouped into pseudo-bands with the same total occupation, having widths of around a few level separations and separated by the order of the Fermi energy (which is usually much greater than the level spacing). The addition of the charging term will shift non-neutral bands up linearly in the charging energy with an increasing slope for charged configurations. One should be able to compensate for these changes by using a gate or bias voltage, as discussed below.

One of the simplest ways of taking the charging energy into consideration in the regime of strong charging energy and weak lead binding is within a multi-electron master equation approach. The correctness of the method requires that the effects of broadening be negligible, which is quite generally not true, but the approximation nevertheless provides some insight into the behavior of nanometric devices when charging has been accounted for, particularly when a nonzero bias has been applied. This formalism is statistical only, and unfortunately this makes discussion of perfect devices that send every electron to the right place more difficult than it is in the single-electron formalism. Alternatively, one can utilize a perturbation treatment in the lead-device coupling, recently suggested by König and Gefen for an AB mesoscopic ring; however, this approach is limited to off-resonant transport only.

Before we proceed to discuss the calculation of the current within the multi-electron master equation approach, we briefly digress to the matter of estimating the charging energy and its variation with ring geometry. Calculating the energy needed to (uniformly) charge a ring or torus is an elementary electrostatics problem, though not one to which an analytical solution exists to our knowledge. The calculation below provides a qualitative estimate of the magnitude of the charging energies and the numbers should not be taken as being accurate to within more than an order of magnitude.

The details of the calculation are as follows. As a model we used for simplicity a cylinder of radius $a$ and height $h$ with a cylindrical hole of radius $b$ through its axis (see inset of Fig. 6 for a sketch of the model). The potential $\phi(r, z)$ at $r$ and $z$ can be obtained following a standard procedure and is given by:

$$\Phi(r, z) = \int_b^{a} \int_0^{2\pi} \int_0^{h} \int_0^{h'} \frac{e}{\rho} \cdot r' \cdot \phi' \cdot \varphi' \cdot (z' - z)^2$$

The charging energy for a single electron may then be found by setting $\rho$ to $\frac{e}{\hbar \pi (a^2 - b^2)}$ inside the ring and zero otherwise:

$$U_0 = \frac{1}{2} \int d \rho \varphi(r, z)$$

The charging energy for $N$ electrons is then given by $U_0(N - N_0)^2$ where $N_0$ is the neutral number of electrons.

In Fig. 6 we plot the charging energy of a nanometric ring as a function of the cylinder radius $a$ and height $h$. For typical rings considered in this work with a radius of $a = 5nm$, a height of $h = 1nm$, and $b = a + h = 6nm$ the charging energy is of the order of $U_0 < \frac{1}{2} eV$. For smaller dimensions the charging energy increases considerably and can exceed several electron volts for sub-nanometer rings.

B. Calculating the Spin Current

If one neglects spin-dependent multi-electron effects, then it is formally straightforward to construct from the set of one-electron Hamiltonian and spin eigenfunctions an anti-symmetric basis of multi-electron wavefunctions:

$$\Psi_{n_1 n_2 \ldots n_i} = A_{1 \ldots i} \prod_{n_i = 1}^{\varphi_{n_i}}$$
Here $A_{1,...,i}$ is the antisymmetrization operator and the states are identified by their (spin-dependent) level occupations $n_i$ (0 or 1 for fermions). Using this antisymmetric multi-electron wavefunction we can uniquely

\[
\langle \varphi_i | G | \varphi_j \rangle = g_{ij},
\]

\[
\langle \Psi_{n_1 n_2 ... n_M} | G | \Psi_{n_1 n_2 ... n_M} \rangle = \sum_j g_{jj} n_j
\]

\[
\langle \Psi_{n_1 n_2 ... n_M} | G | \Psi_{n_1 n_2 ... n_M} \rangle = g_{kk}
\]

\[
\langle \Psi_{n_1 n_2 ... n_{i\beta} ... n_M} | G | \Psi_{n_1 n_2 ... n_{i\beta} ... n_M} \rangle = g_{kk} \delta_{n_i - n_i'} - \delta_{n_k - n_k'} + 1
\]

\[
\langle \Psi_{n_1 n_2 ... n_{i\beta} ... n_M} | G | \Psi_{n_1 n_2 ... n_{i\beta} ... n_M} \rangle = 0,
\]

where $M$ is the number of single-electron levels taken into account, and $n_i' = \lfloor 1 - n_i \rfloor$. Multi-electron effects will be considered only in the form of charging energy. Since these values will be used in a rate-process calculation rather than a full quantum formulation, constructing the multi-electron states themselves is actually redundant, and Eqs. (20a) (20c) along with the single particle data will provide all the necessary information.

In order to perform a master equation based estimation of the current, the transfer rates between different multi-electron states must first be calculated. We will assume that only levels near the Fermi energy will take part in conduction. This implies that the $N$ levels closest to the Fermi energy will be used to construct the $2^N$ multi-electron states themselves using Eq. (20b), with the additional charging term $U_0(N - N_0)^2$.

The transfer rate through lead $\ell$ between two multi-electron states is given by:

\[
R_{\ell,\alpha \rightarrow \beta} = \frac{\Gamma_{\ell,\alpha \beta}}{\hbar} Q_{\alpha \beta},
\]

where multi-electron states by the Greek indices $\alpha \equiv \{ n_1^{(\alpha)}, n_2^{(\alpha)} \cdots n_N^{(\alpha)} \}$ and $\beta \equiv \{ n_1^{(\beta)}, n_2^{(\beta)} \cdots n_N^{(\beta)} \}$. Single electronic levels are labeled by the indices $i$ and $j$. We also define the total transfer rate summed over all leads:

\[
R_{\alpha \rightarrow \beta} = \sum_{\ell} R_{\ell,\alpha \rightarrow \beta}.
\]

In the above equations, $\Gamma_{\ell,\alpha \beta}$ is related to the the imaginary part of the self-energy in the single electron picture. To lowest order, $\Gamma_{\ell,\alpha \beta} = \gamma_{\ell,ii}$ if the two multi-electron states differ only by the occupation of level $i$, $\Gamma_{\ell,\alpha \beta} = \gamma_{\ell,ij}$ if they differ only by $n_i$ and $n_j$ and $n_i - n_j = 1$. Otherwise $\Gamma_{\ell,\alpha \beta} = 0$. This follows from the Slater-Condon rules (cf., Eqs. 20b and 20d), $\gamma_{\ell,ij}$ is the matrix element of the imaginary part of the self-energy. $Q_{\alpha \beta}$ in

\[
Q_{\alpha \beta} = \begin{cases} f(\epsilon_\alpha - \epsilon_\beta - \mu) & N_\alpha > N_\beta, \\ 1 - f(\epsilon_\alpha - \epsilon_\beta - \mu) & N_\alpha < N_\beta, \\ 1 & N_\alpha = N_\beta, \end{cases}
\]

Eq. [21] is related to the Fermi-Dirac function, $f(\epsilon)$:

\[
Q_{\alpha \beta} = \begin{cases} f(\epsilon_\alpha - \epsilon_\beta - \mu) & N_\alpha > N_\beta, \\ 1 - f(\epsilon_\alpha - \epsilon_\beta - \mu) & N_\alpha < N_\beta, \\ 1 & N_\alpha = N_\beta, \end{cases}
\]

where $N_\alpha = \sum_i n_i^{(\alpha)}$ is the number of electrons in state $\alpha$.

With the rates known, the linear master equation system can be written as the condition for steady-state:

\[
\sum_{\beta} R_{\alpha \rightarrow \beta} P_\alpha - \sum_{\beta} R_{\beta \rightarrow \alpha} P_\beta = 0,
\]

where $P_\alpha$ is the probability that the system is in a multi-electron state $\alpha$. Once the steady-state occupation probabilities have been solved for the current can be expressed as:

\[
I_\ell = -e \sum_{\alpha \beta} R_{\ell,\alpha \rightarrow \beta} P_\alpha s_{\alpha \beta},
\]

Intuitively, this expression states that current flows out of lead $\ell$ whenever an electron flows from it into the device, with the inverse also true. Following a similar line of physical reasoning leads to an expression for spin-polarized current: up or down current flows out of lead $\ell$ whenever an up or down electron flows from it into the device. Assuming no coupling between levels with different spin, the spin-dependent current is given by:

\[
I_{\ell,\uparrow(\downarrow)} = -e \sum_{\alpha \beta} R_{\ell,\alpha \rightarrow \beta} P_\alpha s_{\uparrow(\downarrow)\alpha \beta},
\]
and

$$s_{\uparrow(\downarrow)}^{(\alpha,\beta)} = \begin{cases} +1(0) & S_\alpha < S_\beta \land N_\alpha < N_\beta, \\ 0(\pm 1) & S_\alpha > S_\beta \land N_\alpha < N_\beta, \\ 0(-1) & S_\alpha < S_\beta \land N_\alpha > N_\beta, \\ -1(0) & S_\alpha > S_\beta \land N_\alpha > N_\beta. \end{cases}$$

(28)

Here, $S_\alpha = \sum_i s_i^{(\alpha)}$ where $s_i^{(\alpha)} = \pm 1$ for spin up or down, respectively.

Peaks in the differential conduction as a function of the bias voltage $V_b$ can be expected whenever there exists a difference in energy between two states differing in their number of electrons by one, which is occupied in one lead but not the other, i.e., when the conduction window grows to contain a spectral line. This is why levels spaced more than about $eV_b + k_BT$ from the Fermi level should not take part in conduction within this formalism: transfer through them involves electrons or holes (referring here to level vacancies) not present in the leads.

C. Spin Filter

A filtering device remains straightforward in this formalism, and the previous discussion in subsection IIB pertains to it as well. Several differences are nevertheless evident. In the single electron formalism a device will conduct at zero bias when the chemical potential coincides with an energy level on the device. In the master equation formalism this still happens when the charging energy is zero. Under such conditions, the difference in energy between the neutral state and the first charged state always equals the Fermi energy, and this difference is obviously contained in the zero-bias conduction window.

In the presence of charging, the picture is somewhat more involved. The same states discussed above are now shifted differently by charging since they have a different number of electrons. For the sake of clarity, we will first consider the simplest case where only 2 levels and thus 4 states are included. We assume that the system can have $N_0 - 1$ electrons, $N_0$ electrons, or $N_0 + 1$ electrons with energies $U_0$, $\epsilon + g\mu_0 B$, and $2\epsilon + U_0$, respectively ($\epsilon$ is the single electron level energy). Furthermore, $N_0$ is the neutral occupation and we assume it is such that one of the aforementioned levels is occupied when the system is neutral. The value of these energies was calculated by taking the single electron level $\epsilon$ and adding charging terms ($U_0$ for both $N_0 - 1$ and $N_0 + 1$) and magnetic field splittings (for the $N_0$ state).

When the chemical potential in one of the leads equals the energy difference between multi-electron states a peak will appear in the differential conductance. For the case described here when the bias voltage falls symmetrically on both junctions, conduction peaks will occur when the energy difference between two multi-electron states of different occupations is equal to the value of the lead Fermi energy combined with half the applied bias voltage. Taking the Fermi energy as that of the lower of the two single electron conduction levels, the conduction peaks are expected to appear at $eV_b = 2U_0$ and $eV_b = 2(U_0 + 2g\mu_0 B)$ (these peaks are doubly degenerate).

Typical behavior of a spin filter device is illustrated in Fig. 7, where $U_0 = \frac{1}{10}\text{eV}$, $\epsilon = 1\text{eV}$, and $T = \frac{1}{2}\text{K}$. On the left, surface plots of the current as a function of the gate and bias voltages are shown (panels (a)-(d)). Upper
panels show the current of spin up (left) and spin down (right) electrons for a wide range of gate and bias voltages. Note how current steps (or conduction peaks) occur whenever the conduction window cuts across a difference between state energies as discussed above. For instance, with no gating transitions with $N → N ± 1$ occur simultaneously at $eV_n = 2U_0$, and with a gate voltage of $±U_0$ the first step occurs at zero bias. On the scale of the plots shown in panels (a) and (b), the currents for the two different spins are almost indistinguishable. The difference in current between the two polarization can be seen in panels (c) and (d), where we zoom in on a specific area of the line described by $eV_0 = U_0 − eV_0/2$, where $V_0$ is the gate voltage. Here, one spin starts to flow at slightly lower bias voltages, the current rising with the voltage to a high peak and then falling back down as the other spin begins to flow as well. A plane cut through this surface is shown in panel (e), where it is clearly demonstrated how by tuning the voltages a spin filter can still be constructed when charging is taken into account. A symmetric filter for the opposite spin type is found when inverting the gate voltage. As can be seen in the figure, the first conduction peak occurs at $eV_n = 2(U_0 − 2g\mu_0B)$ and the second occurs at $eV_n = 2U_0$. As a result of the broken symmetry, these peaks are non-degenerate and correspond to transition with $N → N − 1$. The other two peaks corresponding to $N → N + 1$ will occur at a significantly higher bias voltage. This is in contrast to the simple example where only 4 states were considered, as describe above, where the current peaks are degenerate and appear simultaneously.

One of the interesting features shown in Fig. 7 is the negative differential spin conductance of the spin up electron (panel (g)). This is explained by the sudden drop in the population of the $|01\rangle$ state (corresponding to a conduction electron of spin up) as the change in chemical potentials begins to allow the population of state $|10\rangle$ (corresponding to a conduction electron of spin down). This population switching is reminiscent of the nonmonotonic change of occupation in two electrostatically coupled single-level quantum dots. The state populations are shown in panel (h) of Fig. 7. The drop in the spin up state population is correlated with the occurrence of negative differential spin conductance (the conductance is normalized to the maximal value). This can be explained in the following way: the current for each spin is determined by the product of the probability that the system is in state $\alpha$ and the rate of transitions between state $\alpha$ and state $|00\rangle$ (which is the state with no conducting electrons), where $\alpha = |10\rangle$ for spin up conducting electron and $|01\rangle$ for spin down conducting electron. We therefore expect that at chemical potentials where the relevant Fermi functions and hence the rates are nearly constant, the current will be approximately proportional to the population.

D. Spin Splitter

The discussion of the spin splitter within the multi-electron master equation is more involved than the spin filter and requires at least 4 levels (16 states). We assume that at $N_0$ (the neutral occupation) two of the four conduction levels are occupied. Having previously established a way in which single electron levels suitable for spin splitting can be found on a ring, we focus on the effects of charging. First, and for the moment neglecting charging energies, we assume that we have found two levels with energies $\epsilon ± \Delta$ that are spin degenerate without Zeeman splitting, with the following properties: they should be adjacent (other than perhaps for non-transmissive levels, which will not qualitatively affect our results), and should both be coupled symmetrically to the input lead, while each is coupled to a single different output lead. Other levels should be at least $2\Delta$ away in energy. All couplings are taken to be the same and diagonal couplings are ignored. Accounting for Zeeman splitting will simply shift each of these levels in energy by $±g\mu_0B$ without modifying the coupling. As before for the single electron picture, the coupling between levels with different spins is neglected. An example of such a case is discussed above in Sec. III C where $\Delta$ is given in terms of the ring level spacing (c.f., Eq. [15] and Fig. 4).

FIG. 8: A spin-splitter made from two levels 1eV apart where $\Delta = \frac{1}{2}eV$, each of which is further split by the Zeeman energy at 10$T$. The upper diagrams show the conductance through the two output channels when the charging energy is ignored and the lower diagrams shows the conductance of the two output channels at a charging energy of $U_0 = 0.6eV$. We have adjusted the gate potential so that in both cases the current rises at $V_n \approx 1$Volt and at this value only one spin conducts through each lead.

Setting the chemical potential of the input lead to $\mu_1 = \epsilon + eV_0$, and both output chemical potentials to $\mu_0 = \epsilon − eV_0$, it should now be clear under both formalisms that without charging energy a perfect spin splitter exists at $eV_n = 2\Delta − 2g\mu_0B$, as shown in the upper
panels of Fig. 8. In the single electron picture this happens as two levels with opposite spins and different lead bindings enter the conduction window. In the multi-electron master equation this happens when the difference in energy between \( N = N_0 \) states and \( N = N_0 \pm 1 \) states enters the conduction window. These two conditions are equivalent for \( U_0 = 0 \).

Including charging effects will cause the \( N = N_0 \pm 1 \) states to move up in energy by \( U_0 \). This has the effect of repositioning the conductance peaks at \( eV_h = 2\Delta \pm 2\mu_B B \pm U_0 \). If \( U_0 \) is of the order of the Zeeman term then the charging energy will interfere with the structure that allowed our spin splitter for \( U_0 = 0 \), since the energy difference between states that differ by one electron of both spin up and spin down can become similar. On the other hand, if the charging energy is increased further, the transitions \( N = N_0 \rightarrow N + 1 \) and \( N = N_0 \leftrightarrow N - 1 \) become distinguishable in energy. One way to take advantage of this is to apply a gate potential in order to shift the state energies by \(-U_0\) so that only the \( N = N_0 \leftrightarrow N + 1 \) transitions are in the conduction window (new transitions may appear at similar energies if other levels are too close, but since level spacing for a ring generally increases with energy we can neglect such contributions). Once again at \( eV_h = 2\Delta - 2\mu_B B \) perfect splitting occurs, though at a smaller total current, since fewer state transitions are involved. This is apparent in the lower panels of Fig. 8.

V. CONCLUSIONS AND DIRECTIONS FOR FURTHER STUDY

We have investigated several ways in which rings or ring-like structures with a radius of the order of nanometers, coupled to metallic leads, might be used to construct simple spin-sensitive devices. We have focused only on the Zeeman splitting to differentiate between spins, since it is always present while other effects commonly utilized in microscopic structures, such as the Rashba and Dresselhaus effects, are generally absent in molecular/nanometric structures. We believe the niche this work occupies in the search for nanospintronic devices is yet unexplored: very small coherent structures and weak spin-dependent effects have not received much attention, despite their formal simplicity and significance. What we have shown here is that although building devices under the burden of such limitations is difficult, it is possible. Considering the scientific and technological benefits of such devices we believe it will also be worthwhile.

The basic calculations we have performed are enough to point out to the right direction as to the desired properties of molecule-sized Aharonov-Bohm spin devices and the conditions which their desired operation might be observed. A similar methodology can be applied to more complex devices or sets of devices. The actual devices specifically discussed here were two of the most basic - a spin-filter and a spin-splitter or Stern-Gerlach device. However, the conclusions drawn and principles laid out may easily be extended to many interesting systems, from quantum gates (since in theory we can use interference devices to perform general unitary transformations between input and output gates) to molecular memory (since an electron trapped on the device will modify its electrical properties and thus may be detectable at a later time).

Several complementary methods were utilized during the course of this investigation: first, a simple single-electron, analytical model in which the parameter space can easily be explored, and thus basic intuition about the system can be gained. Second, a tight-binding nonequilibrium Green’s function treatment, which incorporates a more realistic physical structure that can be compared directly with experimental data, but still assume independent electrons, was applied to similar systems studied within the simple analytical model. Finally, a multi-electron master equation approach that can be used to examine many-particle effects was grafted onto the tight-binding results. Here, we choose to focus on electric charging, since its effects are energetically dominant.

Our analysis of the filter, meant to be a test-case, was based on the study of the spinless case where it was possible to create very narrow conduction peaks near zero magnetic field by combining weak device/leads couplings and a gate voltage to shift the conduction resonances to \( B = 0 \). Since the Zeeman splitting depends linearly on the magnetic field, a spin filter always requires a finite magnetic field, at least high enough to separate the spin conduction peaks in energy by more than \( k_B T \). Control over the position of the spin-dependent conduction peaks and their widths can be achieved by carefully adjusting the kinetic phase

\[
\phi_{k}^{11} = \frac{\pi R}{\hbar} \sqrt{2m^*} \left( \epsilon + \frac{2e\hbar\mu_B B}{\sqrt{2m^*}} \phi_m \right)
\]

and the coupling between the device and the leads, respectively. Unlike the spinless case where \( \phi_k = \frac{\pi R}{\hbar} \sqrt{2m^*} \), the kinetic phase is now a function of the magnetic flux itself, and thus, the conduction is not a simple periodic function of the parameters. High efficiency spin filters are constructed at the highest magnetic field possible, where spin-dependent effects are strongest, and flexibility is gained by selecting materials or structures with high effective mass (or large ring size, which is not desirable). Charging effects do not drastically modify this picture since spin selectivity depends only the Zeeman term and the application of a gate voltage can compensate for the charging energy itself. However, charging leads to a breaking of symmetries and as a result to negative differential spin-conduction.

The physics of the spin splitter device is similar to that described above with the added complication that differentiated control of the spin-dependent wavefunction at the different leads is required. Within the single electron picture, a spin splitter device may operate when two spin-degenerate levels exist such that one transmits through one lead only and the other through the other lead only. Furthermore, the level separation should equal
effects of spin-orbit coupling and the local spin density
electron structure description. One such is through the
Zeeman splitting which require a more elaborate elec-
tronic systems, we see ways of enhancing the effect of
particular, drawing from the study of crystals and meso-
important set of directions to continue along. In par-
ticipants can reduce the overall current through the device. However,
charging does not abolish the general picture and the
application of a gate potential can be used to overcome
most of its effects.

For both prototype devices we have shown how the
limits of lead-coupling, system geometry and tempera-
ture at which one might expect to see the desired effects
can easily be estimated, as well as several ways in which
one can implement specific behaviors by systematically
finding parameters at which they occur, either exactly
or approximately. We have also shown that even within
the parameters space of a device formed by a single ring
with two or three leads, nontrivial behaviors with useful
properties take place. In at least two instances, we have
made a case that our findings are physical rather than
a mathematical peculiarity of some model by reproduc-
ing them under different assumptions and formalisms.
We have found an interesting example of negative dif-
ferential conductance for a spin polarized current with
a simple explanation. Finally, we have described what
is, to our knowledge, the smallest Stern-Gerlach appara-
tus ever reasonably conceived of, and one which is fully
switchable in directionality at constant magnetic field by
the application of an external electric field.

Despite their simplicity, the calculations reported here
provide several novel predictions that seem to be model
independent. Nevertheless, more refined models are an
important set of directions to continue along. In par-
cular, drawing from the study of crystals and meso-
scopic systems, we see ways of enhancing the effect of
Zeeman splitting which require a more elaborate elec-
tronic structure description. One such is through the
effects of spin-orbit coupling and the local spin density
exchange-correlation energy, both of which have been
used to explain the giant spin g-factor enhancement that
has been observed in mesoscopic structures under certain
conditions. It is quite possible that the same effect can
be recreated with discrete levels, although this probably
requires that only a small number of levels be occupied
(corresponding once again to very low electron densities).

While we have studied only two- and three-terminal
devices, with only one injective terminal, it is reasonable
that four terminal devices will also be of interest as coher-
ent quantum gates. One of the reasons we have found it
worthwhile to draw attention to the importance of many-
particle effects is our hope that in time-dependent cal-
culations they may be used to create sequential logical
behavior without sacrificing coherence - for instance, an
electron may only be able to enter the device through one
lead, but when it enters it opens up another lead, and the
inflow of another electron causes both to be discharged
through a third lead - a sort of sequential AND gate, two
of which could form a true AND gate. Of course, to con-
sider useful computation it is necessary to model entire
networks of such gates, where input electrons enter on
one set of leads, propagate throughout the network and
leave it on another set of leads. Such a network presents
new delocalized challenges unless devices are somehow
coupled in such a way that no inter-device interference
takes place. However, even if the devices within the net-
work are all "imperfect" devices that only approximate
logic gates, as long as every device has independently ad-
justable parameters (like a gate voltage) such a network
forms a fascinating basis for a model of a quantum neu-
ral networks. These and related issues are still open for
future study.

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