Spintronics birefringence with an extended molecular loop-wire or spiral coupling

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A ring with spin-orbit effects coupled to a conducting wire is shown to exhibit a phase delay which is spin dependent. The key is that the coupling of the ring to the wire is over an extended spatial range and not just along a single point; this breaks the symmetry and makes the ring states couple differently to forward and backward moving wire states. This results, for properly injected spin states, in a spin-flipping probability which is dependent on the energy of the injected electron and can therefore be easily controlled. Several systems are presented and shown to exhibit this effect including the basic ring which couples to a wire as well as a ring which mediates between two wires, and a spiral between two wires. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2126664]

I. INTRODUCTION

One of the basic problems in the molecular and nanoelectronics is spintronics, i.e., how to control electron-spin degrees of freedom in solids or in artificially manufactured semiconductor nanodevices.[1–5] These devices include quantum dots, wires, nanotubes, and even molecules. Recently, new man-made semiconductor nanoscale systems have emerged—nanorings on Si.[6,7] Due to the nontrivial topology of these systems they possess interesting quantum mechanical and magnetic properties.[8–12] Similarly, ring-based molecular systems exhibit interesting interference effects.[13,14] In this paper we study how ring-shaped nanodevices and molecules could be useful in electron-spin control.

Probably the most natural tool for controlling spins is the magnetic field. The use of the magnetic field gives rise to such mechanisms as Aharonov-Bohm and Zeeman effects. Different ways for exploiting the magnetic field in spintronics have been proposed and could be found in recent reviews.[1,2,5] Magnetic fields have the advantage that they are externally applied,[12] however, the effect of the magnetic field is proportional to the flux so that it is difficult to apply it for molecular systems which have inherently small areas, or to very small nanosystems.

Another physical tool which can control spins is the spin-orbit effect. Spin-orbit effects are often quite strong for molecular systems (with splittings of more than an eV for heavier atoms) but are trickier to apply. A possible device we propose in this paper has the ring (Aharonov-Bohm) topology and also has an inherent spin-orbit interaction. The reason why such a device might be interesting for spintronics is that the spin is coupled to the molecular angular momentum l_z, an internal quantum number of the device. If the device, in addition, has the $l_z \rightarrow -l_z$ symmetry broken (e.g., geometrically wise), the spin degeneracy would also be broken.

The simplest geometry that breaks $l_z \rightarrow -l_z$ symmetry would be to connect a heavy atom to two ligands at an angle; then the electron which passes through the heavy atom from one ligand to the other one will travel in a definite direction (clockwise or counterclockwise, depending on the geometry) so that the $l_z \rightarrow -l_z$ symmetry is broken and due to the spin-orbit interaction different spins experience different interactions.

One feature of the spin-orbit interaction, however, is that the phase delay it induces does not depend on the energy. (Semiclassically, this is because the spin-orbit interaction is proportional to the velocity of the particle, and the integral of this interaction is just proportional to the length of the trajectory of the electron). This feature is advantageous in some systems, since it makes the spin-orbit effects less sensitive to perturbations. However, this also makes it more challenging to gate the effect. Further, in the process of electron propagation a single element, which does not retain the electron for a considerable period of time, may not always be strong enough to cause a considerable spin rotation. The loop cavity geometry we propose in this paper allows both to effectively extend and to gate the spin-orbit interaction.

The suggested geometry is equivalent to a cavity and makes it possible to use either (or both) magnetic fields or spin-orbit couplings to affect the spin. In its simplest version, the geometry is just a loop weakly coupled to a wire, with slightly different geometries presented later. The key, however, is to have the wire and the electron couple along a range not just at a single point. By having this extended coupling the $l_z \rightarrow -l_z$ symmetry of the loop is broken, so that different magnetic spins experience slightly different phases along their motion. Qualitatively, an incoming wave on the wire would couple differently to a clockwise and to an anticlockwise state on the cavity. Therefore, different orbital angular momenta experience different interactions, leading through the spin-orbit interaction to a spin-dependent motion.

Heuristically, the difference in interaction has two effects: one is that at sufficiently strong energy differences one spin projection could be blocked. This, however, requires the

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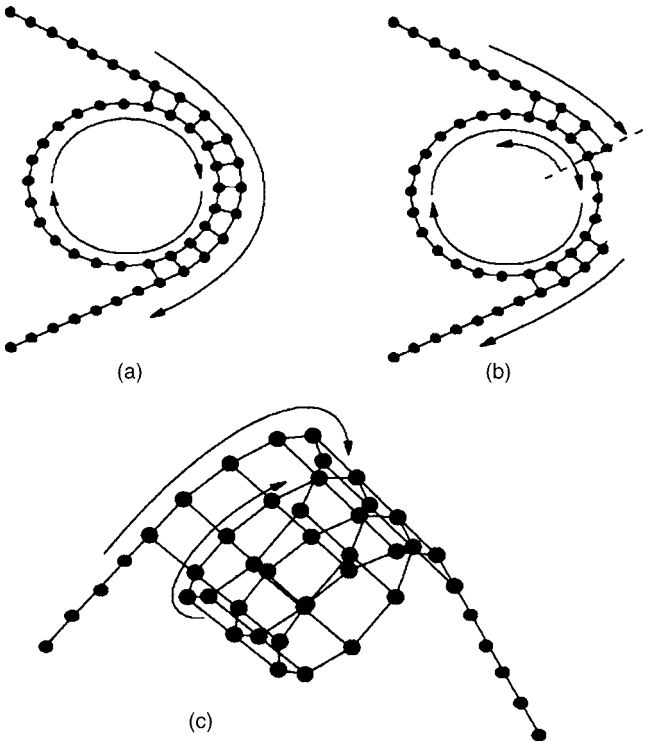

FIG. 1. Three geometries studied. (a) The wire-loop geometry, (b) the wire-loop-wire geometry, and (c) wire-spiral-wire geometry.

breaking of the planar symmetry so it would not appear in the systems considered here which are all effectively planar. The other effect is more subtle and is important even for small energy-splitting and planar systems; it is the different phase velocity of the two spins. This is completely analogous to optical birefringence, where light that passes through an uniaxial crystal experiences different phase delays for different polarizations. This phase delay can be properly manipulated to cause spin rotation.

In this paper we explore the implications of the spin birefringence in three Huckel models. Section II introduces the models: wire loop, a similar wire-loop-wire model in which the coupling between the two leads is only through the loop, and a spiral which connects two wires. It then discusses briefly several physical systems where this effect can be used, including silicon rings, ringlike molecules, and nanotubes. Section III presents the results. Discussion of the relevance of these effects to specific spintronics systems, using either spin-orbit interactions or also magnetic field (in combination with gating effects) follows in Sec. IV.

II. SYSTEMS

The numerical models we consider should possess the two significant properties discussed in Sec. I. The first is the broken $l_z \rightarrow -l_z$ symmetry, or broken "orbit symmetry" as one might call it, and the second is the spin-orbit coupling interaction on the ring. We propose three such models (Fig. 1).

A. Wire-loop geometry

The first model is of a wire in conjunction with a loop [Fig. 1(a)]. The current flows along the wire, and the wire is coupled along a range to the loop. A Huckel Hamiltonian is used; the coupling between different sites is then summarized as

$$H_{I,J} = \begin{cases} 2\beta, & I = J \\ -\beta, & I,J \text{ connected, both on wire} \\ -\beta e^{ig_{I,J}}, & I,J \text{ connected, both on loop} \\ -\gamma, & I,J \text{ connected; } I \text{ on loop,} \\ & J \text{ on wire or vice versa.} \end{cases} \quad (1)$$

.

In detail, γ is a weak coupling which connects wire and loop sites that are neighboring (are in parallel sites). Also, all sites on the wire are connected by a kinetic coupling (2β on the diagonal and $-\beta$ on the off diagonal); on the loop there is a similar coupling but it is modulated by the spin-orbit effect. The modulation is through a factor which we label as

$$g_{I,J} = \begin{cases} \pm \dfrac{a}{\beta} u(\max(I,J)) s_z, & I = J \pm 1 \\ 0 & \text{otherwise,} \end{cases} \quad (2)$$

where s_z is the spin projection along the z axis, i.e., the axis perpendicular to the loop, a is the strength of the spin-orbit coupling, and the function u defines the specific model as discussed below.

There are several options for the spin-orbit coupling. The first one, considered here, is an equal spin-dependent coupling along the loop. In this case, $u(I)=1$. Such an extended spin-orbit coupling can be induced if the loop is surrounding a single atom or a magnetic cluster which induces a magnetic coupling. Such a coupling would typically be weak, since it acts essentially by a remote-atom-effect, i.e., the electrons which experience the spin-orbit effect would be far from the heavy atom. The second option (not considered here) for the wire-loop coupling is a loop which includes in it a single heavy atom which is connected in an angle. It is straightforward to prove that such a connection provides a phase delay which depends on the spin; formally, then, $u(I)$ would be 1 at the site of the single heavy atom and zero otherwise. Finally, a more general option is a loop in which several heavy atoms are placed.

In this geometry, the most important aspect is that the wire-loop coupling is extended over several sites. Therefore, an incoming wave on the wire couples mostly to the eigenstates on the loop with clockwise rotation. Mathematically, an incoming wave would have an eigenstate of the form $\exp(ikj)$, where j is the site on the wire and k is the associated (dimensionless) wavevector; on the loop it would be, if the loop is essentially undistorted, $\exp(im\varphi)$, where m is the eigenvalue of l_z and φ is the angle spanned by the site. Obviously, if there is a match between the arguments of the two eigenstates ($m\phi \sim kj$) there would be good coupling, and the coupling would only be effective for m of a single sign (for an incoming wave).

The wire-loop geometry is reminiscent of a racetrack, since the wave that comes into the loop (the "track") joins into the loop gradually, making it go only in one direction

(counterclockwise or clockwise), unlike a regular T junction which distributes the particles symmetrically in both directions.

B. Wire-loop-wire and wire-spiral-wire general

The second geometry is a wire which connects to a loop and that connects in turn to another wire. If only a single loop is used, this geometry [Fig. 1(b)] is reminiscent of a "packman," but several loops can tie together to form a spiral which self-connects. Here, coupling must occur through the loop. The Hamiltonian used is the same as Eq. (1), except that now two (unconnected) wires are employed. The qualitative difference from the wire-loop geometry is that the outgoing wave inevitably passes through the loop.

Note the similarity to a third option, a wire which connects to a regular spiral. The main difference between a wire-spiral-wire and a wire-loop-wire options is that a regular spiral is open, so that the wave cannot do a full revolution in it.

C. Chemical relevance

There are several systems which can be envisioned built on this model. Detailed simulations will be presented in future papers, but for reference we briefly mention several options. The first is, as mentioned in the introduction, nanorings. Here, the idea would be to coat the nanorings, from inside, with a transition-metal compound. The transition-metal compound would induce a spin-orbit rotation along the ring. In this case the relevant energy scale would be in the sub-eV regime.

A more immediately feasible approach would be to use a molecular system. Here, a transition metal would be surrounded by a ring of hydrocarbons. There are many other variants on this; even a single transition-metal atom connected at an angle to two ligands which are then connected below would act as a small birefringent loop. Here, the energy scale would be similar to that in the simulations, a few eV; since the gating will be done by tuning the working Fermi level or the injection energy, the gating voltages will also be a few volts.

Finally, the spiral simulation is obviously reminiscent of a directed nanotube, built around a scaffold of transition metals, though other geometries are also feasible.

Similar geometries would be feasible for use with magnetic fields, which would necessitate, however, large fields or the use of larger rings.

III. METHODOLOGY

We calculate both the total reaction probability for going from the one lead to the other, as well as the scattering probability to start from a specific spin and end up, on the other wire, with a different spin.

The total reaction probability is a function of the incoming spin. All the systems we consider are planar or quasiplanar and therefore S_z would be approximately conserved, but the scattering phase is spin dependent. Therefore, the Hamiltonian is just labeled by the value of $2S_z$.

The total reaction probability is obtained from the flux-flux reaction rate,[15]

$$N(E) = \mathrm{Tr}(\Gamma_L G \Gamma_R G^\dagger), \tag{3}$$

where[14]

$$G = \frac{1}{(E - H + i\Gamma_L + i\Gamma_R)} \tag{4}$$

is the Green's function in the presence of the absorbing potentials Γ_L and Γ_R, which are applied at both edges of the wire. The more interesting information, however, is in the S matrix. We calculate that from the expression for the full wave function in terms of the absorbing potential[16]

$$\psi = G\Gamma_L \eta_L, \tag{5}$$

where η_L is the free incoming wave in the left lead, i.e.,

$$\eta_L(j) = \frac{1}{\sqrt{v}} \exp(ikj), \tag{6}$$

where the momentum on the lead and its velocity are defined from the energy in the Huckel model as

$$k = a \cos\left(1 - \frac{E}{2\beta}\right), \quad \nu = 2\beta \sin(k) \tag{7}$$

Then, the S matrix is obtained from the overlap of the full wave function with the analogous free wave function from the right lead[15,16]

$$T_{Sz} = \langle \eta_R^* | \Gamma_R | \psi \rangle = \langle \eta_R^* | \Gamma_R G \Gamma_L | \eta_L \rangle, \tag{8}$$

where we denote the explicit dependence of the transition amplitude on S_z.

The T matrix determines the phase delay experienced by each spin. The interesting aspect is that, if the initial spin is polarized, the final spin would be polarized too but with a possibly different polarization. Specifically, define the spin-up ("+") and spin-down ("−") components to be relative to the z axis, which is by definition the axis perpendicular to the plane. Write then the initial spin as a combination of these two states

$$\lambda_{\mathrm{in}} = a_+ |+\rangle + a_- |-\rangle. \tag{9}$$

Then the outgoing spin would be, in an obvious notation,

$$\chi_{\mathrm{out}} = T_+ a_+ |+\rangle + T_- a_- |-\rangle = T_+ \left(a_+ |+\rangle + \frac{T_-}{T_+} a_- |-\rangle \right). \tag{10}$$

Therefore, the spin would be rotated. For simplicity, we assume that the incoming electrons are prepolarized along the positive x axis, so that their spin component would be $\chi_{\mathrm{in}} = (|+\rangle + |-\rangle)/\sqrt{2}$; one measurement of the device is then the "flipping" amplitude (denoted by f), i.e., the component along the negative x axis,

$$f = \frac{(T_+ - T_-)}{2}, \tag{11}$$

as well as the probability for spin flip $|f|^2$.

The devices we propose are based on the effect of different phase delays during the propagation for different spins. Therefore, it can operate only if the incoming electrons are polarized and the polarization is in plane. If the

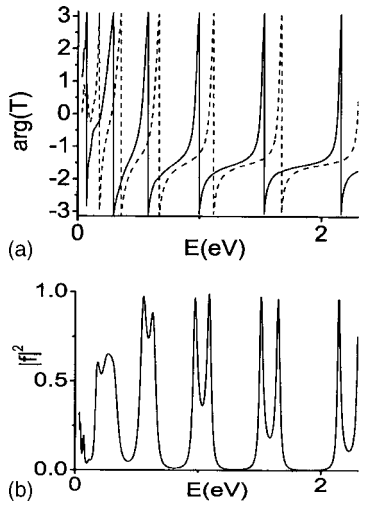

FIG. 2. Results for wire-loop geometry: (a) The phase of the transmission amplitude for spin up (solid line) and spin down (dashed line). The transmission probability is identical for the two spins (b) spin-"flip" probability $|f|^2$ (see Eq. (11)).

incoming electrons are not polarized the effect of spin rotation will be smeared due to statistical averaging over the polarization direction.

IV. RESULTS

All simulations were done with a reasonable value of the kinetic energy constant in the Huckel Hamiltonian for organic and chemical systems, $\beta=2.7$ eV. We used parabolic absorbing potentials which extend through the last 50 sites of the outer ends of the wires. Also, a weak wire-loop coupling was used; for the wire-loop simulations we took $\gamma=0.2$ eV.

The first simulations were for a model wire-loop system (Sec. II A), in which we applied a uniform spin-orbit coupling throughout the loop, with a spin-orbit coupling strength, for each site, of $a=0.02$ eV. The loop has then 40 sites. The number of sites in the wire just has to be large enough for convergence, but, to avoid any convergence issues, we took a total of 200 wire sites (more than needed). The loop wire coupling extended over ten sites.

Figure 2 shows the main results of the loop-wire simulations. We first show in Fig. 2(a) the phase of the transmission amplitude for the two spins. Near specific resonance energies the phases vary rapidly with energy. Therefore, the spin-flip probability, which is related to the difference in the transmission phases, show marked energy dependence near the specific resonances [Fig. 2(b)].

Qualitatively, the results of Fig. 2 can be understood as follows. The transmission pattern in Fig. 2 shows clear Fano resonances associated with the electron states on the loop. In the absence of the spin-orbit coupling the electron states are the orbital states with the quantized angular momentum number, $m=0,\pm1,\pm2\ldots$. The states with opposite electron rotations ($m=\pm|m|$) are twofold degenerate. In the effective-mass approximation the energies of the states are simply proportional to the angular momentum quantum number squared,

$$E_m=\alpha m^2,$$

where α is the parameter depending on the electron effective mass and the physical size of the loop.

When one turns on the spin-orbit coupling the degeneracy is lifted for an electron with a given spin direction. The energy of the states becomes

$$E_m=\alpha m^2+\eta s m,$$

where η is a constant characterizing the strength of the spin-orbit interaction, and related to the parameters in the Hamiltonian.

In the wire-loop model the electrons passing by the loop are mostly coupled to the loop states with angular momentum quantum numbers of a specific sign. In other words, only the loop states in which the electrons rotate clockwise are coupled to the electrons going from the upper part of the wire to the lower one [see Fig. 1(a)]. Thereby, only these states would result in the Fano resonances in the complex amplitude.

For electrons with the opposite spin direction these states are going to be at different energies due to spin-orbit coupling. This means that Fano resonances are going to appear at different energies for the two opposite spins. For instance, the nearby resonances for two spins, which correspond to the loop state with angular momentum $m>0$, are going to be at energies

$$E_m=\begin{cases}\alpha m^2+\eta|s|m, & s=\frac{1}{2}\\ \alpha m^2-\eta|s|m, & s=-\frac{1}{2}.\end{cases} \tag{12}$$

The separation of the resonances for the two spins is clearly seen in Fig. 2(a), where we give the phase of the complex transmission amplitude for the two spins as a function of electron energy.

However, the phases of the transmission pattern are not identical, so the flipping amplitude (Eq. (11)) would be nonzero. The transmission coefficients of both spins enter equivalently, so that both types of resonances (resonances for both spins) contribute to it. Therefore, the flipping amplitude has Fano resonances at energies of both spin-up and spin-down loop states [see Fig. 2].

The discussion of the wire-loop system is seemingly valid only for weak wire-lead coupling. However, the unidirectionality would be valid even for quite strong coupling, since to any perturbation order the coupling of an incoming state on the wire to clockwise states would be different than to anticlockwise states.

We next move to the wire-loop-wire system, which is shown in Fig. 3 to have a different profile of the flipping probability. Specifically, Fig. 3 presents the transmission phase and flipping probability for both spins in a wire-loop-wire system. The parameters used for this figure are somewhat different from the wire loop; a coupling strength of $\gamma=0.15$ eV, and a spin-orbit coupling of 0.015 eV; each wire couples to the loop along ten points.

The wire-loop-wire geometry (Sec. II A) does not reveal the simple physics of the loop wire. The reason is that the upper wire [see Fig. 1(b)], as well as the lower wire, have a boundary (indicated by the blue dashed line). On this bound-

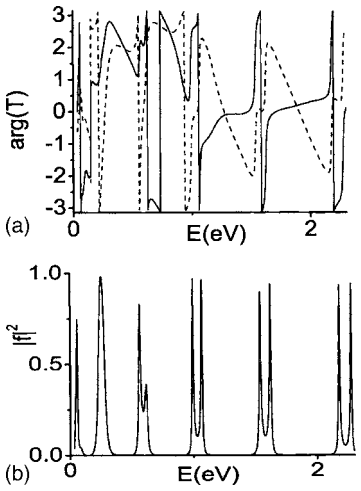

FIG. 3. The same as Fig. 2 for wire-loop-wire geometry.

ary an incoming electron can rebound backwards. Therefore, the electron is going to be coupled not only to the "clockwise" loop states ($m>0$) but also to the "counterclockwise" states ($m<0$). Both spins would have the Fano resonances at the both types of energy levels Eq. (12).

Another interesting issue is the transmission probability in the two models. The transmission probability $|T_+|^2$ for both models is given in Fig. 4. This probability is independent of spin. This is due to the symmetry of the Hamiltonian under inversion coupled with a spin change. Therefore, if an up electron was going backwards along the wire it would couple to the same angular states that a down electron would going forward; but since the transmission probability is independent of the spatial direction in this two-channel model, it follows that these probabilities cannot depend on spin.

A very interesting outcome and quite general result is that for the wire-loop geometry the transmission probability is almost unity for most of the energy range. Heuristically, the reason is that reflection is a much more "violent" event than phase change, which is cumulative. More mathematically, referring to Fig. 1(a), a wave which impinges on the device from the top lead couples only to clockwise rotating waves. These waves do not couple to outgoing states on the top lead, only to outgoing states on the bottom lead, and therefore the transmission probability is close to unity over a large energy range. (At low energies, the coupling length, ten grid points, is not sufficiently large compared to the de Broglie wavelength so that the coupling is less unidirectional and the overall transmission is therefore less than unity.)

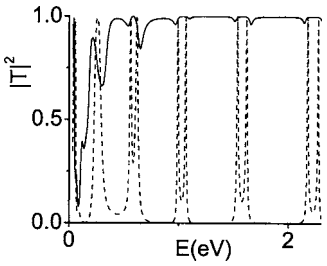

FIG. 4. The transmission coefficient probability $|T|^2$, which is spin independent, for the wire-loop (solid line) and wire-loop-wire (dashed line) models.

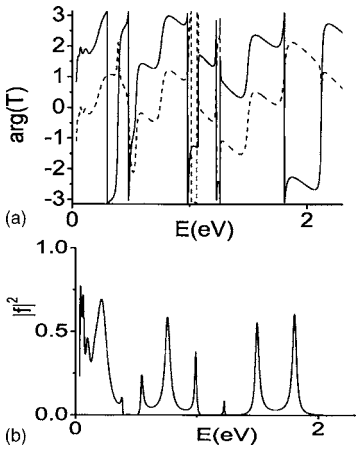

FIG. 5. Similar to Figs. 2 and 3 for the spiral geometry.

The transmission coefficient $T_+ \approx e^{i\varphi}$ is therefore characterized mainly by the "transmission phase," φ (in analogy with the scattering phase in the scattering problem in two dimensions (2D) and three dimensions 3D). The "transmission phase" characterizes the average "time" the electrons spend on the loop, so that at resonances it has singularities (see Fig. 2 for the phase variation); at the same time, even for resonances, the transmission probability would be close to unity because the rapid variation is of the (cumulative) phase.

In contrary, in the wire-loop-wire problem the transmission probability is almost zero unless the electron is at resonance with one of the loop states. This fact is also easy understandable. A probable route for an electron just entered the loop-wire junction is to stumble on the upper wire [blue dashed line in Fig. 1(b)] and be reflected back into the upper wire. Therefore, the transmission probability in the wire-loop-wire model has a pronounced peak structure. In addition, the singularities for both spins, appearing at similar energies, compensate each other to some extent.

The results for the spiral model are given in Fig. 5. The parameters used here are a loop-wire coupling of 0.2 eV and a stronger spin-orbit coupling of 0.1 eV; the two spirals are each connected over five sites. The stronger coupling is needed since electrons do not rotate several times as the spiral is not closed. The spin-flip probability is now usually below 1, but it is still considerable at some regions so that the phase birefringence induced by the spins is considerable.

V. DISCUSSION

An interesting outcome of the simulations emerges, as mentioned, from comparing Figs. 4 and 2(b): For loop-wire systems, the current is almost energy independent but the flipping probability is strongly dependent on energy. Therefore, the system would play a small differential resistance with a strong differential spin resistance.

Another effect, which we did not consider, is the interaction with magnetic fields and their application with loop-wire systems. Magnetic fields interact in two ways: first, they can couple directly to the spin; in addition, they couple to the orbital angular momentum. Therefore, effectively the magnetic field can induce an additional spin-orbital momentum

coupling which was not considered here but will be examined in future publications. The key point using either spin-orbit or magnetic effects is that the coupling or the wire(s) is not symmetric, since the wire and the loop are not at right angles.

There are several ways to use the spin-orbit effects with systems suggested here for practical application. The simplest approach would be to polarize the incoming spin of the electron (using, e.g., a permanently magnetized lead), inject the electrons at specific energies, and then use a gating voltage to modulate the incoming electron internal energy. This will lead to a flipping probability which is easily modulated.

Another possibility is to modulate directly the spin-orbit interaction. This would be possible if the spin-orbit effect is affected by an electronic change (as would be, for example, if it acts by a remote heavy-atom effect) so that it would be affected by light or a chemical change.

To go beyond this, it would be interesting to consider a wire interacting with several consecutive rings. This would lead, presumably, to a sharper band structure which is more easily controlled. Alternately, an effect which we did not consider is nonplanar geometries. Even the spiral we studied is associated with a completely planar Hamiltonian. A true single-handed spiral (left or right handed) could lead to spin rotation rather than just phase delay and flipping.

The phase delay between spin-up and spin-down electrons has other important ramifications. If it could be modulated, then a spin modulator would emerge; this is in complete analogy to electroptic modulators. The modulation would be by light, gating, or even a chemical change. An interesting use of such modulators would be to measure the polarization amount of the incoming electrons.

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