Electrical Manipulation of Nanomagnets

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We demonstrate that it is possible to manipulate the magnetic coupling between two nanomagnets by means of an ac electric field. In the scheme suggested, the magnetic coupling is mediated by a magnetic particle that is in contact with both nanomagnets via tunnel barriers. The time-dependent electric field is applied so that the height of first one barrier then the other is suppressed in an alternating fashion. We show that the result is a pumping of magnetization from one nanomagnet to the other through the mediating particle. The dynamics of the magnetization of the mediating particle allows the coupling to be switched between being ferromagnetic and being antiferromagnetic.

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The sensitivity of electron transport to the spin degree of freedom brings new possibilities for implementing device functions in electronics. As a result, the field of spintronics is developing rapidly. The giant magnetoresistance [1] is a striking example of a spin-dependent transport phenomenon that has already found important applications in computer hardware. More fundamental ideas for using spin in order to realize devices that can store and process quantum information are now under intensive discussion in the literature [2,3].

Manipulation of the electron spin is possible only if one is able to control the magnetization of the magnetic materials that are necessarily part of any spintronics device. In nanoscale devices a fundamental obstacle to achieving the required level of control appears because the magnetic fields used to control the magnetization cannot be localized on the nanometer length scale. This is in sharp contrast to the electric fields used in modern nanoelectronics based on single-electron devices [4]. The problem of how to selectively control the magnetization of a nanomagnet has therefore become crucial for building nanoscale spintronics devices. Using electric rather than magnetic fields to manipulate nanomagnets could, if it works, be a way out of this "nonlocality trap." A natural way to perform such a control is to employ the indirect exchange interaction between nanomagnets. This interaction is induced by the conduction electrons, which obviously can be controlled electrically via the wave functions of the electrons that transfer magnetic polarization between nanomagnets [5–7]. Such a transfer is determined by the interference pattern produced by different electronic waves and is therefore crucially affected by any kind of structural material disorder and by the detailed interface geometry on an atomic scale. As a result the phenomenon becomes very sensitive to fluctuations and noise in the system.

The main idea of this Letter is to explore a novel type of magnetic coupling where magnetization is transferred through a "time domain" rather than through a "spatial domain." This may be the case if the magnetic coupling between two nanomagnets is due to a small movable magnetic particle — a mediator — that in effect "pumps" magnetization from one nanomagnet to the other. Since the magnetization is carried by the mediator, one may maintain a "delay line" with the possibility to control the magnitude and the orientation of the magnetization to be transferred. This type of manipulation can indeed be achieved electrically by means of the exchange interaction, which is essentially of electrostatic origin. Below we propose a new method for electrically controlling the magnetization of nanoscale magnetic matter.

Figure 1 shows a sketch of the model system to be considered. It contains two single-domain nanomagnets with magnetic moments $\mathbf{M}_L$ and $\mathbf{M}_R$. They are both coupled by the exchange interaction extending through a time-dependent tunnel barrier to a magnetic cluster or

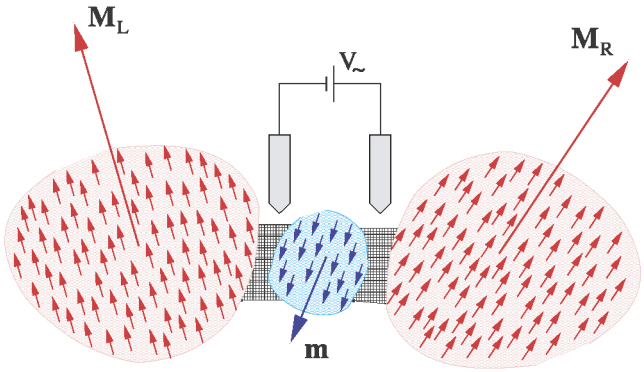

FIG. 1 (color online). Schematic diagram of the system discussed in the text. Single-domain magnetic grains with magnetic moments $\mathbf{M}_L$ and $\mathbf{M}_R$ are coupled by a magnetic cluster — a mediator — with magnetic moment $\mathbf{m}$. Gate electrodes induce ac electric fields that are concentrated to the areas between the grains and the cluster. These fields control the heights of the tunnel barriers and therefore affect the exchange coupling between the different components of the system.

molecule with magnetic moment $\mathbf{m}$ [8]. An indirect exchange interaction between the two nanomagnets is mediated by the cluster/molecule which acts as a magnetic weak link between the magnets. An important feature is that the exchange coupling between the mediating magnetic moment and the magnetic leads is controlled by the heights of the tunnel barriers that separate the nanomagnets and the cluster. We will show that by applying to the tunnel barriers an electric field that varies periodically in time, the nature of the weak link can be transformed so as to be a mediator of either a ferromagnetic or an antiferromagnetic coupling.

The most interesting regime of operation occurs if the exchange coupling between mediator and leads is made to have a time dependence that corresponds to a sequential coupling of the mediator to first one of the magnetic leads and then to the other, in a periodically repeating pattern. In this case three temporally separated contributions to the resulting coupling between the two leads can be identified: (1) polarization of the mediator by one of the magnetic leads (while the mediator is essentially decoupled from the other lead); (2) domination of the internal dynamics of the free mediator (when the mediator is decoupled from both leads); (3) transfer of the induced magnetic polarization on the mediator to the second lead (while decoupled from the first). While steps (1) and (3) are essential for the device operation, what happens during step (2) is not qualitatively important. For simplicity we will omit this step. Under these conditions the time evolution of $\mathbf{m}$ can be thought of as being due to a sequence of "scattering events." A single event changes the magnetic moment of the mediator by $\Delta\mathbf{m}$. Because of the conservation of magnetic momentum, the magnetic moment of the lead also changes. Therefore, one can view the entire process as a mediator-assisted flow of magnetic polarization between the leads. This flow, giving rise to a synchronized evolution of the magnetization in the leads, establishes an effective coupling between them.

Now let us demonstrate qualitatively how one, by manipulating the polarization flow using a time-dependent electric field, can affect the magnetic coupling between the leads. Since $M \gg m$ the dynamics of the magnetization in the leads is much slower than the dynamics of the magnetic moment (spin) of the mediator. When considering the dynamics of the mediator magnetization, one can therefore to a first approximation neglect the variation of $\mathbf{M}$ altogether. Then, the time-dependent exchange coupling of the mediator to the leads will result in a periodically oscillating effective magnetic field acting on the magnetic moment of the mediator. Any weak relaxation effects present will bring the mediator magnetization $\mathbf{m}(t)$ into a periodic regime for which $\mathbf{m}(t) = \mathbf{m}(t + 2T)$. In this regime the magnetic moment of the mediator changes from, say, $\mathbf{m}_1$ to $\mathbf{m}_2$ during the first half-period T when the mediator cluster is coupled to, say, the left lead. During the second half-period, when it is coupled to the right nanomagnet, the reverse change takes place (from $\mathbf{m}_2$ to $\mathbf{m}_1$). During the time it is coupled to a lead, the mediator — under the influence of the effective magnetic field that points in a fixed direction — performs a rotation around an axis parallel to the magnetization of the lead (see Fig. 2).

The total angle of rotation $\phi_\alpha = g\bar{J}_\alpha M_\alpha T$ ($\alpha = L, R$), after the mediator has been magnetically coupled to the lead during one contact, depends on the average exchange coupling strength $\bar{J}_\alpha$ and the effective coupling time T ($g = 2\mu/\hbar$, μ is the Bohr magneton). One finds that, in the symmetric case ($\phi_L = \phi_R = \phi$) to be considered here, the vector $\Delta\mathbf{m}$ is perpendicular to the plane spanned by $\mathbf{M}_L$ and $\mathbf{M}_R$ (the x-y plane). The flow of polarized magnetization will result in a rotation of $\mathbf{M}_\alpha$ around an axis parallel to the vector $\mathbf{M}_L + \mathbf{M}_R$ (the x axis). This is a signature of the existence of an effective magnetic field $\mathbf{h}$ directed along that axis (see Fig. 2). Relaxation processes that are inevitably present will tend to orient the magnetization of the lead along this field. Let us suppose that the rotation angle $\phi = \phi_0$ is much smaller than 2π. Under this condition the vectors $\mathbf{m}_{1,2}$ will be oriented almost along the bisector of the angle between $\mathbf{M}_L$ and $\mathbf{M}_R$, and

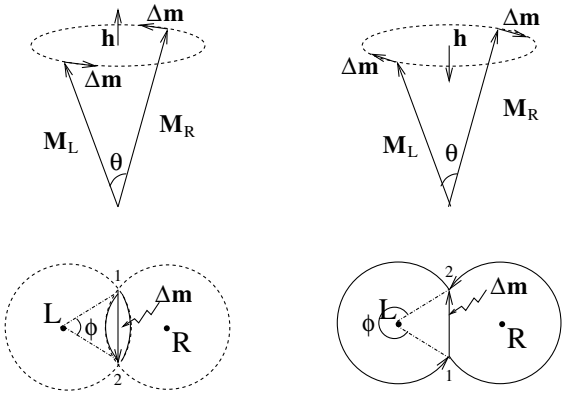

FIG. 2. Illustration of the periodic regime of the molecular spin dynamics in which magnetization flows in one direction between the two nanomagnets of Fig. 1. The bottom parts represent the periodic dynamics of the projection of the molecular spin on the plane perpendicular to the vector $\mathbf{M}_L + \mathbf{M}_R$. Points L and R indicate the axes aligned with the vectors $\mathbf{M}_L$ and $\mathbf{M}_R$, respectively. If the molecular cluster is coupled to a nanomagnet, its spin performs counterclockwise rotation around axis L or R (depending on which nanomagnet it interacts with). The circles schematically represent the trajectories which are traced out by the end of the vector $\mathbf{m}$. The angle of rotation ϕ depends on the length of time the cluster is coupled to the nanomagnets and on the magnitude of the exchange coupling, which is controlled by the amplitude of the alternating field applied to the gate electrodes. In the periodic regime of the molecular spin evolution the spin periodically oscillates between points 1 and 2. During one oscillation period a magnetic moment $\Delta\mathbf{m}$ is transferred from one nanomagnet to the other, making the nanomagnet magnetizations rotate around the axis $\mathbf{M}_L + \mathbf{M}_R$ (upper part).

therefore $\mathbf{h}$ will be directed along the vector $\mathbf{M}_L + \mathbf{M}_R$. In such a case the magnetic moments of the leads, since they tend to be oriented along the effective magnetic field, will be ferromagnetically ordered ($\theta = 0$).

Now let us consider the regime that appears when the rotation angle is $2\pi - \phi_0$. One finds that if a rotation by an angle ϕ_0 around some axis gives rise to a change of magnetic moment from $\mathbf{m}_1$ to $\mathbf{m}_2$, the rotation around the same axis by the angle $2\pi - \phi_0$ will transform $\mathbf{m}_2$ into $\mathbf{m}_1$. Therefore, a periodic evolution of $\mathbf{m}(t)$ will be established where during the first half-period (when the mediator is coupled to the right lead) its moment changes from $\mathbf{m}_2$ to $\mathbf{m}_1$ and vice versa during the second half-period. So we will have the same magnetic flow, but in the opposite direction. From this analysis one can immediately conclude that the effective magnetic fields at ϕ_0 and at $2\pi - \phi_0$ will be pointing in opposite directions. Consequently, at $\phi = 2\pi - \phi_0$ the $\mathbf{h}$ should be antiparallel to the vector $\mathbf{M}_L + \mathbf{M}_R$ making the ferromagnetic ordering unstable. Below we will show that if $\phi > \pi$ the system is actually antiferromagnetically ordered. Therefore, by tuning the rotation angle ϕ — which depends on the amplitude and frequency of the alternating electric field — one can create a transition from ferromagnetic to antiferromagnetic coupling between the magnetization in the leads.

For a quantitative discussion of the phenomena outlined above, we will use the Landau-Lifshits equations:

$$\frac{1}{g}\frac{d\mathbf{m}}{dt} = \left(\frac{\partial W}{\partial \mathbf{m}} \times \mathbf{m}\right) + \frac{\beta}{m}\left[\mathbf{m} \times \left(\mathbf{m} \times \frac{\partial W}{\partial \mathbf{m}}\right)\right], \quad (1)$$

$$\frac{1}{g}\frac{d\mathbf{M}_\alpha}{dt} = \left(\mathbf{M}_\alpha \times \frac{\partial W}{\partial \mathbf{M}_\alpha}\right) + \frac{\beta}{M}\left[\mathbf{M}_\alpha \times \left(\mathbf{M}_\alpha \times \frac{\partial W}{\partial \mathbf{M}_\alpha}\right)\right].$$

Here $M = |\mathbf{M}|$, $m = |\mathbf{m}|$ and the magnetic energy W of the system can be expressed as

$$W = -\sum_{\alpha=L,R} J_\alpha(t)(\mathbf{M}_\alpha \cdot \mathbf{m}), \quad (2)$$

where $J_\alpha(t)$ describes a periodic time-dependent exchange coupling (with period $2T$) between mediator and magnetic leads. In this work we take $J_{L,R} = \bar{J}[1 \pm \alpha(t)]/2$ with $\alpha(t) = \mathrm{sgn}(\sin \pi t/T)$. The terms proportional to β in Eq. (1) describe relaxation with relative characteristic frequency β. In what follows we will assume $\beta \ll 1$ (the value of β varies from 0.5 to 0.005 between different magnetic materials [9]). In this case the dissipation only slightly affects the magnetization dynamics and nontrivial regimes can be expected. If $M \gg m$, the dynamics of the molecular spin is much faster than the dynamics of the magnetization of the leads, and one can use the adiabatic approximation to analyze the behavior of the system. To do this we will calculate $\mathbf{m}(t)$ under the assumption that the magnetization of the leads is fixed and then substitute it into Eq. (2). Averaging over the fast oscillation one gets the equation $\frac{1}{g}\frac{d\mathbf{M}_\alpha}{dt} = (\bar{\mathbf{h}}_\alpha \times \mathbf{M}_\alpha) + \frac{\beta}{\mathbf{M}}[\mathbf{M}_\alpha \times (\mathbf{M}_\alpha \times \bar{\mathbf{h}}_\alpha)]$ for $\mathbf{M}_\alpha$, where the effective magnetic fields $\bar{\mathbf{h}}_\alpha$ are given by the relation $\bar{\mathbf{h}}_\alpha \equiv \frac{1}{2T}\int_0^{2T} dt J_\alpha(t)\mathbf{m}(t)$. Therefore the dynamics of the magnetization of the leads is controlled by the average spin polarization of the mediator when it is coupled to the lead. Integrating Eq. (1) over one period one finds $\mathbf{m}(2T) - \mathbf{m}(0) = gT\{(\mathbf{M}_L \times \bar{\mathbf{h}}_L) + (\mathbf{M}_R \times \bar{\mathbf{h}}_R)\}$. This implies that for the case of periodic evolution, when $\mathbf{m}(2T) = \mathbf{m}(0)$, the average fields $\bar{\mathbf{h}}_L$ obey the relations $(\mathbf{M}_L \times \bar{\mathbf{h}}_L) = -(\mathbf{M}_R \times \bar{\mathbf{h}}_L)$. Taking the scalar product of this relation with $\mathbf{M}_\alpha$ one can easily find that the projection of $\bar{\mathbf{h}}_\alpha$ on the axis perpendicular to the plane spanned by $\mathbf{M}_L$ and $\mathbf{M}_R$ plane is equal to zero in the periodic regime. As a result, $\bar{\mathbf{h}}_\alpha$ may be presented as a linear combination of magnetizations $A\mathbf{M}_\alpha + \bar{J}\mathbf{M}_\beta$, where the coefficient $\bar{J}$ is some function of the angle θ between the vectors $\mathbf{M}_L$ and $\mathbf{M}_R$. One can represent the magnetic fields through an effective interlead interaction energy $\mathcal{W}$ as $\mathbf{h}_\alpha = -\delta\mathcal{W}/\delta\mathbf{M}_\alpha$.

The structure of the effective potential $\mathcal{W}$ controls the type (ferromagnetic or antiferromagnetic) of the interaction between the nanomagnets. Since $\mathcal{W}$ depends only on the angle θ, and hence can be represented as a function of the scalar product $\mathbf{M}_L \cdot \mathbf{M}_R$, one can prove the relations $\mathbf{e}_z \cdot (\mathbf{M}_L \times \mathbf{h}_L) = -[\mathbf{e}_z \cdot (\mathbf{M}_R \times \mathbf{h}_R)] = \partial\mathcal{W}(\theta)/\partial\theta$ (the z axis is chosen parallel to $\mathbf{M}_R \times \mathbf{M}_L$). Using these relations one gets the equation for the time evolution of the angle θ as

$$\frac{M}{g}\frac{d\theta}{dt} = \beta\frac{\partial\mathcal{W}}{\partial\theta}. \quad (3)$$

On the other hand, multiplying Eq. (1) by $\mathbf{e}_z$ and integrating over the first half-period $(0, T)$ — when the molecular spin is coupled to the left lead — one finds that $\Delta m_z/T = g[\partial\mathcal{W}(\theta)]/\partial\theta$. On combining this result with Eq. (3) one finds that the time evolution of the angle θ is determined by the equation

$$\frac{1}{T}\Delta m_z = \left(\frac{M}{\beta}\right)\frac{d\theta}{dt}. \quad (4)$$

The quantity $\Delta m_z/T \equiv \bar{j}$ has a simple physical interpretation; it gives the average flow of the z component of magnetization between the leads, mediated by the periodic evolution of the mediator magnetization. As a result a mutual rotation, with frequency $\Omega = \bar{j}/M$, of the vectors $\mathbf{M}_\alpha$ around the x axis takes place.

In order to decribe the fast dynamics of the molecular spin $\mathbf{m}$, it is convenient to use a matrix representation. Let $\hat{\boldsymbol{\rho}}$ be a (2×2) matrix for which $\mathrm{Tr}\hat{\boldsymbol{\rho}} = 0$, $\mathrm{Tr}\hat{\boldsymbol{\sigma}}_i\hat{\boldsymbol{\rho}} = 2m_i/m$, if $i = x, y, z$, and σ_i are Pauli matrices. Equation (1) can now conveniently be written as

$$\dot{\hat{\boldsymbol{\rho}}} = -i[\hat{H}(t), \hat{\boldsymbol{\rho}}] - \beta[\hat{\boldsymbol{\rho}}, [\hat{\boldsymbol{\rho}}, \hat{H}(t)]], \quad (5)$$

where $\hat{H}(t) = (1/2)\sum_{\alpha=L,R} gMJ_\alpha(t)e^{i\theta\hat{\sigma}_z/4}\hat{\sigma}_x e^{-i\theta\hat{\sigma}_z/4}$. Since the "Hamiltonian" $\hat{H}(t)$ is a periodic function of

time, the solution of Eq. (5) can be expressed in terms of "quasienergy" states $|t, \pm\rangle$ defined by the equations $i(d/dt)|t, \pm\rangle = \hat{H}(t)|t, \pm\rangle$, where $|t + 2NT, \pm\rangle = e^{\pm i\lambda N}|t, \pm\rangle$. In this representation the matrix $\hat{\rho}$ has the form

$$\hat{\rho} = \rho(t)(|t, +\rangle\langle +, t| - |t, -\rangle\langle -, t|) + \tau(t)|t, +\rangle\langle -, t| + \tau^*(t)|t, -\rangle\langle +, t| \quad (6)$$

with $\rho^2 + |\tau|^2 = 1$. For $\beta = 0$ Eq. (6) is a solution of Eq. (5) with ρ and τ being time independent. If $\beta \ll 1$ the coefficients ρ and τ are slow functions of time. Their time evolution can be found by substituting for $\hat{\rho}(t)$ in Eq. (5) and averaging over one period. In the case when the mediator is never coupled to both leads simultaneously, the states $|t, \pm\rangle$ may be found exactly. As a result one finds that $\rho(t)$ obeys the equation $d\rho/dt = \beta g\bar{J}M(1 - \rho^2) \times C(\theta, \phi)\cos\phi/2$, where $C(\theta, \phi) = (1 - \sin^2\phi/2\cos\theta/2)^{-1/2}\cos\theta/2$. It follows that the molecular spin relaxes to the periodic regime of evolution ($|\tau| \to 0$) and that in this regime $\rho = \mathrm{sgn}(\cos\phi/2)$.

Now we can calculate $\bar{j} = m(2T)^{-1}\mathrm{Tr}\hat{\sigma}_z[\hat{\rho}(T) - \hat{\rho}(0)]$. Making use of Eq. (4) one obtains the equation

$$\frac{d\theta}{dt} = -\beta T^{-1}\frac{m}{M}\mathrm{sgn}(\cos\phi/2)B(\theta, \phi)\sin\theta \quad (7)$$

for the time evolution of the angle θ. Here $B(\theta, \phi) = \sin^2(\phi)/|\sin\lambda|$. From Eq. (7) we conclude that the relative magnetization of the leads depends on ϕ—the precession angle of the molecular spin during the time it is coupled to the lead. If $2n\pi < \phi < (2n + 1)\pi$ the mediated exchange interaction imposes a ferromagnetic ordering between the single-domain nanomagnets. If on the other hand $(2n - 1)\pi < \phi < 2n\pi$, the angle θ increases and the system tends to establish an antiferromagnetic ordering. However, our analysis based on the adiabatic approximation breaks down for the narrow θ interval defined by $|\theta - \pi| \leq m/M \ll 1$. Since $\phi = gMJ_0e^AT$, where the exponent $A = V/V_0$ is proportional to the amplitude of the ac potential applied to the tunnel barriers, one can switch the magnetic ordering of the nanomagnets between being ferromagnetic to being antiferromagnetic by varying the amplitude of the electric field (or the oscillation period).

As one can see from Eq. (7), the switching rate $d\theta/dt$ is proportional to the relaxation parameter β, as expected for a magnetic system. Although dissipation is essential for the mediator dynamics to be periodical, the effective exchange energy $\mathcal{W}$ in this regime [see Eq. (4)] does not depend on β to leading order. The sign and magnitude of $\mathcal{W}$ are actually controlled by the angle of the mediator spin precession as seen from Fig. 2. Still, a small but finite dissipation does lead to a correction to $\mathcal{W}$ that is proportional to β. A more detailed analysis shows that this correction has a ferromagnetic character for all values of the period of oscillations (amplitude of electric field) and that it provides a ferromagnetic coupling of the leads if dissipation is strong ($\beta \sim 1$). Therefore, in principle, manipulation of the magnetic coupling as described above is possible only if β is lower than some critical value $\beta_c \sim 1$. However, to the best of our knowledge, β is generally much less than unity for real magnetic materials.

In conclusion, we suggest a new type of voltage-controlled exchange coupling between nanomagnets that interact via a small magnetic particle separated from the nanomagnets by tunnel barriers. We have demonstrated that by using a time-dependent electric field to periodically suppress first one then the other tunnel barrier, both a ferromagnetic and an antiferromagnetic ordering of the two nanomagnets can be achieved. It is furthermore possible to switch from one type of ordering to the other by changing the amplitude or frequency of the applied ac voltage. As an alternative to using an ac voltage, sequential coupling between the mediator and the two nanomagnets could be achieved by means of a movable mediator that "shuttles" magnetization between the nanomagnets. This method would be analogous to the setup used in recent experiments [10,11], where shuttling of electric charge [12] has been observed.

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