

Pumping *ac* Josephson current in the Single Molecular Magnets by spin nutation

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We demonstrate that an *ac* Josephson current is pumped through the Single Molecular Magnets (SMM) by the spin nutation. The spin nutation is generated by applying a time dependent magnetic field to the SMM. We obtain the flowing charge current through the junction by working in the tunneling limit and employing Green's function technique. At the resonance conditions some discontinuities and divergencies are appeared in the normal and Josephson currents, respectively. Such discontinuities and divergencies reveal themselves when the absorbed/emitted energy, owing to the interaction of the quasiparticles with the spin dynamics are in the range of the superconducting gap.

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Quantum pumping is a coherent transport mechanism to produce a charge current in the absence of an external bias voltage by an appropriate periodical variation of the system parameters [1–4]. It has been introduced as a potential way to generate a dissipationless charge current in the nanoelectronic devices [5]. The adiabatic quantum pumping is also a method for generating a dynamically controlled flow of spin-entangled electrons, which is promising because of the vast expertise already available in solid-state electronics [6]. In recent years, electron pumps consisting of different systems such as small semiconductor quantum dots [7–9], carbon nanotube quantum dot [10], one-dimensional interacting Luttinger liquid quantum wire [11], Josephson junctions with half-metallic ferromagnets [12] and diffusive ferromagnets [13], and InAs nanowire embedded in a superconducting quantum interference device (SQUID) [14] have received considerable theoretical and experimental attentions. Several different mechanisms have been proposed to pump charge through such systems, ranging from a low-frequency modulation of gate voltages in combination with the Coulomb blockade to photon-assisted transport.

Magnetic Josephson junctions consisting of Single Molecular Magnets (SMM) and magnetic nanoparticles have recently attracted intense attentions, owing to their applications in molecular spintronics devices [15] and classical [16] and quantum information processing [17]. The small sizes of these junctions are an advantage for their application. Compounds of single molecular magnet class [18, 19] are particularly attractive for application in high-density information storage and quantum computing, due to their long magnetization relaxation time at low temperatures [20, 21]. The rich physics behind the

magnetic behavior produces interesting effects such as negative differential conductance and complete current suppression [22, 23], which could be used in nanoelectronics.

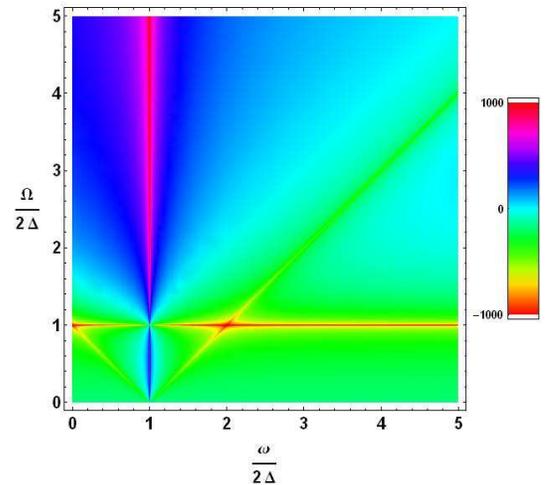


FIG. 1: (Color online) Color map of the amplitude of the *ac* Josephson current \bar{J} as a function of $\frac{\omega}{2\Delta}$ and $\frac{\Omega}{2\Delta}$. At $\frac{\Omega}{2\Delta} \sim 1$ the amplitude goes down to very small values (infinity) for $\frac{\omega}{2\Delta} \neq 1$. At $\frac{\omega}{2\Delta} \sim 1$ the amplitude diverges for $\frac{\Omega}{2\Delta} \neq 1$.

In this letter, we show that the spin nutation of a SMM could pump the charge current through the Josephson junction consisting of the SMM. The spin nutation can be generated by applying an external time dependent magnetic field to the SMM which is a combination of a static and a rotating transverse *rf* fields. We consider a SMM connected to the two spin-singlet superconducting (SSC) leads via tunnel barriers. We investigate the coupling of the spin nutation and Josephson current through the junction. Interplay of the Josephson current and a precessing spin between various types of superconducting leads connected via tunnel barriers has

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been considered by Zhu and Balatsky [24]. The Josephson current through the junction consisting of two SSC leads is not modulated by the spin precession. Whereas, when both superconductors have equal spin triplet pairing state, spin precession causes to modulate the Josephson current with twice the Larmor frequency. In addition, it has been shown that a circularly polarized ac spin current with the Larmor frequency is generated in the SSC leads in response to the spin precession [25].

Working in the tunneling limit and employing normal and anomalous Green's functions we obtain the flowing current through the junction. We have found that the normal current, the current associated to the single particle tunneling, and Josephson current are modulated by spin nutation. In response to the time-dependent boundary conditions induced by spin nutation, ac normal and Josephson currents are pumped in the junction. Some discontinuities and divergencies are appeared in the amplitudes of the ac normal and ac Josephson currents, respectively (Fig. 1). Such discontinuities and divergencies reveal themselves when the absorbed or emitted energy of charge carriers, owing to the interaction with the spin dynamics are in the range of the superconducting gap. At these situations the resonance conditions are fulfilled by the system.

Model - Let us consider the SSC|SMM|SSC Josephson junction. By considering the SMM as a classical spin vector (\mathbf{S}), and applying the time dependent magnetic field $\mathbf{h}(t) = (-h_0 \sin \omega t \sin \Omega t, h_0 \sin \omega t \cos \Omega t, h_z)$ to the SMM, in the absence of the spin relaxation processes the dynamics of the spin is given by:

$$\mathbf{S}(t) = S (\sin \theta(t) \cos \varphi(t), \sin \theta(t) \sin \varphi(t), \cos \theta(t)). \quad (1)$$

Where

$$\varphi(t) = \Omega t, \quad \theta(t) = \theta_0 - \vartheta \cos \omega t, \quad (2)$$

$\Omega = \gamma h_z$ (γ denotes gyromagnetic ratio) is the precession frequency around the z axis and $\theta(t)$ is the time dependent tilt angle (angle between the spin and z axis). The tilt angle oscillates about θ_0 with frequency ω and amplitude $\vartheta = \gamma h_0 / \omega$. (See Fig.(2)) Indeed, this nutational motion of the spin is served as the pumping parameters in the system. Let us consider the following Hamiltonian for the Josephson junction:

$$H(t) = H_L + H_R + H_T(t). \quad (3)$$

The first two terms describe the energy of the left (L) and right (R) spin singlet superconducting leads and are given by

$$H_\alpha = \sum_{k, \sigma = \uparrow, \downarrow} \varepsilon_k c_{\alpha, k, \sigma}^\dagger c_{\alpha, k, \sigma} + \sum_k \left(\Delta_\alpha c_{\alpha, k, \uparrow}^\dagger c_{\alpha, -k, \downarrow} + h.c. \right) \quad (4)$$

where $c_{\alpha, k, \sigma}^\dagger$ ($c_{\alpha, k, \sigma}$) is the creation (annihilation) operator of an electron on the lead $\alpha = L, R$ with momentum

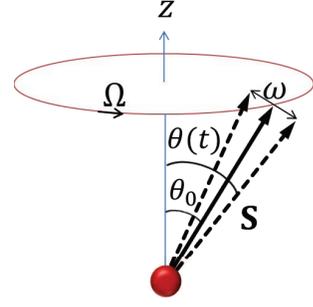


FIG. 2: Schematic representation of SMM spin nutation.

k and spin σ . ε_k is the energy of single conduction electron and $\Delta_\alpha = \Delta e^{i\chi_\alpha}$ is the pair potential in which χ_α is the superconducting phase of the lead α . The two leads are weakly coupled via the tunneling Hamiltonian, $H_T(t)$ which is given by

$$H_T(t) = \sum_{k, k', \sigma \sigma'} \left(c_{R, k, \sigma}^\dagger T_{\sigma \sigma'}(t) c_{L, k', \sigma'} + h.c. \right) \quad (5)$$

$T_{\sigma \sigma'}$ is a component of the following time dependent tunneling matrix

$$\hat{T}(t) = T_0 \hat{\mathbf{1}} + T_S \hat{\mathbf{S}}(t) \cdot \hat{\boldsymbol{\sigma}}, \quad (6)$$

where $\hat{\mathbf{1}}$ is the 2×2 unit matrix, $\hat{\boldsymbol{\sigma}} = (\sigma_x, \sigma_y, \sigma_z)$ is the Pauli matrices and $\hat{\mathbf{S}}(t) = \frac{\mathbf{S}}{|\mathbf{S}|}$ is the unit vector along the SMM spin direction. T_0 is the direct spin independent transmission amplitude and T_S , which is originated from the exchange interaction between the spin of conduction electrons and the localized spin of SMM, indicate the spin dependent transmission amplitude.

In the tunneling limit and zero bias voltage, we could formally separate the current operator at lead α in two parts [26]

$$I_\alpha(t) = I_\alpha^s(t) + I_\alpha^J(t) \quad (7)$$

where $I_\alpha^s(t) = -e \int_{-\infty}^t dt' \langle \langle [A_\alpha(t), A_\alpha^\dagger(t')] \rangle \rangle + h.c.$ and $I_\alpha^J(t) = -e \int_{-\infty}^t dt' \langle \langle [A_\alpha(t), A_\alpha(t')] \rangle \rangle + h.c.$ are the normal current and Josephson current carried by single particles and Cooper pairs, respectively. The operator $A_\alpha(t)$, is given by

$$A_\alpha(t) = \sum_{k, k', \sigma \sigma'} c_{\alpha', k, \sigma}^\dagger(t) T_{\sigma \sigma'}(t) c_{\alpha, k', \sigma'}(t), \quad (8)$$

where $\alpha = L, R$ and $\alpha' = R, L$.

Normal current - In the Following we will show that the spin nutation causes to transfer electrons through the junction and a time-dependent normal current emerges. Let us define the following retarded potential

$$U_{\rho \rho'}^{\sigma \sigma'}(t - t') = -i \Theta(t - t') \langle \langle [a_{k, k'}^{\sigma \sigma'}(t), a_{p, p'}^{\dagger \rho \rho'}(t')] \rangle \rangle, \quad (9)$$

where, $a_{k,k'}^{\sigma\sigma'}(t) = c_{\alpha,k,\sigma}^\dagger(t)c_{\alpha,k',\sigma'}(t)$ and $a_{p,p'}^{\rho\rho'}(t') = c_{\alpha,p,\rho}^\dagger(t')c_{\alpha,p',\rho'}(t')$. Since there is no spin dependent interaction inside the superconducting leads, the retarded potential (9) is independent on spin indices and $U_{\rho\rho'}^{\sigma\sigma'}(t-t') = \delta_{\sigma\rho}\delta_{\sigma'\rho'}U_{ret}(t-t')$. The Fourier transformation of the Matsubara potential, with imaginary time, is defined as [26]

$$\mathcal{U}(i\omega_n) = \frac{1}{\beta} \sum_{iq} g_R(k, iq - i\omega_n) g_L(k', iq), \quad (10)$$

where g_R and g_L are the normal Matsubara Green's functions and ω_n 's indicate the Matsubara frequencies. The retarded potential is obtained from the Matsubara potential by analytical continuation $i\omega_n \rightarrow \alpha + i\eta$ where $\eta \rightarrow 0^+$. Employing Lehman representation we can calculate the retarded potential and obtain the normal current

$$i^s(t) = \Delta T_\perp \left[2T_\perp \mathcal{S} \left(\frac{\Omega}{2\Delta} \right) + T_\parallel \bar{\mathcal{S}} \left(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta} \right) \vartheta \cos \omega t \right], \quad (11)$$

where $i^s = I^s/2\pi e N_L N_R$ and $N_L(N_R)$ is the density of states at the Fermi energy in the left (right) lead. To obtain this equation we have considered $\vartheta/\theta_0 \ll 1$. This approximation is fulfilled by the practical conditions of the system and does not make any restriction on it. The parameters $T_\parallel = T_S \cos \theta_0$ and $T_\perp = T_S \sin \theta_0$ are spin conserving and spin-flip transmission amplitudes, respectively. As it is clearly seen, the normal current strongly depends on the spin-flip transmission amplitude. If T_\perp is zero the normal current vanishes completely. This situation corresponds to the oscillation of the SMM around $\theta_0 = n\pi$ ($n = 0, 1, \dots$). If the spin-flip transmission amplitude is nonzero ($T_\perp \neq 0$), depending on the strength of precession frequency Ω and tilt angle oscillation frequency ω , the single particles could be transferred from one lead to another by absorbing and emitting a quantum of oscillation. In this case the normal current depends on the parameters $\bar{\mathcal{S}}(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta})$ and $\mathcal{S}(\frac{\Omega}{2\Delta})$, which are given by

$$\begin{aligned} \bar{\mathcal{S}} \left(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta} \right) &= 2\mathcal{S} \left(\frac{\omega}{2\Delta} \right) - 2\mathcal{S} \left(\frac{\Omega}{2\Delta} \right) - \mathcal{S} \left(\frac{\Omega+\omega}{2\Delta} \right) \\ &\quad - \mathcal{S} \left(\frac{\Omega-\omega}{2\Delta} \right) - \mathcal{S} \left(\frac{\omega-\Omega}{2\Delta} \right), \end{aligned} \quad (12)$$

$$\mathcal{S}(x) = \Theta(x-1) \left\{ \frac{x^2}{x+1} K(\gamma) - (x+1)[K(\gamma) - E(\gamma)] \right\},$$

where $\gamma = (x-1)/(x+1)$, $K(x)$ and $E(x)$ are the first and second kinds of complete elliptic integrals, respectively. The normal current has *dc* and *ac* parts which both are zero in the absence of the spin precession. The spin precession is sufficient for existence of the *dc* part and it is independent of the tilt angle oscillation. Whereas, the *ac* current is emerged when the tilt angle θ oscillates with a small oscillating amplitude ϑ and the spin conserving transmission amplitude is nonzero. Totally, when $\Omega + \omega$ become larger than 2Δ the normal current is nonzero and

different situations will appear depending on the values of Ω and ω . For $\Omega < 2\Delta$, the *dc* part of the current is zero and three discontinuities appear at points $\Omega + \omega = 2\Delta$, $\omega = 2\Delta$ and $\omega - \Omega = 2\Delta$. Indeed such discontinuities appear in the normal current when the gained or lost energy during the interaction of the single particles with the spin dynamics are almost equal to the superconducting gap. At these points a resonance occurs in the junction. Moreover for $\Omega > 2\Delta$, there is a *dc* normal current in the junction and the discontinuities appear at points $\Omega - \omega = 2\Delta$, $\omega = 2\Delta$ and $\omega - \Omega = 2\Delta$.

Josephson current - To calculate the Josephson current we define a different retarded potential as

$$X_{\rho\rho'}^{\sigma\sigma'}(t-t') = -i\Theta(t-t') \left\langle \left[a_{k,k'}^{\sigma\sigma'}(t), a_{p,p'}^{\rho\rho'}(t') \right] \right\rangle. \quad (13)$$

As in the normal case, in the absence of the spin dependent interactions inside the leads the retarded potential (13) could be written as $X_{\rho\rho'}^{\sigma\sigma'}(t-t') = \sigma\sigma'\delta_{\sigma,-\rho}\delta_{\sigma',-\rho'}X_{ret}(t-t')$, where $\sigma, \sigma' = \pm 1$. The associated Matsubara potential reads

$$\mathcal{X}(i\omega_n) = \frac{1}{\beta} \sum_{iq} \mathcal{F}_R^\dagger(k, iq) \mathcal{F}_L(k', iq - i\omega_n), \quad (14)$$

where \mathcal{F}_R and \mathcal{F}_L are the anomalous Green's functions in the leads [26]. The retarded potential is obtained by analytical continuation. Implementing the real part of the retarded potential

$$\Re \left(\sum_{k,k'} X_{ret}(x) \right) = \begin{cases} \pi N_L N_R \Delta K(x) & x < 1 \\ \pi N_L N_R \frac{\Delta}{x} K(\frac{1}{x}) & x > 1 \end{cases} \quad (15)$$

and defining $\mathcal{J}(x) = \Re \left(\sum_{k,k'} X_{ret}(x) \right) / \pi N_L N_R$, the final form of the Josephson current is given by:

$$\begin{aligned} i^J(t) &= [2(T_0^2 - T_\parallel^2)\mathcal{J}(0) - 2T_\perp^2\mathcal{J}(\frac{\Omega}{2\Delta}) \\ &\quad - T_\perp T_\parallel \bar{\mathcal{J}}(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta}) \vartheta \cos \omega t] \sin \chi \end{aligned} \quad (16)$$

where $i^J = I^J/2\pi e N_L N_R$, $\chi = \chi_R - \chi_L$ is the phase difference between superconducting leads and

$$\begin{aligned} \bar{\mathcal{J}} \left(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta} \right) &= 2\mathcal{J}(0) - 2\mathcal{J} \left(\frac{\Omega}{2\Delta} \right) + 2\mathcal{J} \left(\frac{\omega}{2\Delta} \right) \\ &\quad - \mathcal{J} \left(\frac{\omega+\Omega}{2\Delta} \right) - \mathcal{J} \left(\frac{\omega-\Omega}{2\Delta} \right). \end{aligned} \quad (17)$$

In the special case of $\vartheta = 0$, which corresponds to the $h_0 = 0$, the spin has only a precessing motion about the z axis without tilt angle oscillation. In this case the *dc* Josephson current $i^J = 2[(T_0^2 - T_\parallel^2)\mathcal{J}(0) - T_\perp^2\mathcal{J}(\frac{\Omega}{2\Delta})] \sin \chi$ is generated through the junction. Indeed, the interaction of the quasiparticles with the spin precession affects the *dc* Josephson current and causes to appear a divergence at $\Omega = 2\Delta$, when the quantum of precession is close to the superconducting gap.

For $\vartheta \neq 0$ and $\theta_0 \neq \frac{n\pi}{2}$, depending on the values of $\bar{\mathcal{J}}(\frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta})$, an *ac* Josephson current is generated

through the junction. This modulation of the Josephson current can be used for single spin detection. At $\Omega = 0$, the parameter $\tilde{\mathcal{J}}$ vanishes for arbitrary values of ω and the Josephson current $i^J = 2(T_0^2 - T_{\parallel}^2)\mathcal{J}(0)\sin\chi$ is time-independent. In Fig. (1), we have shown the density plot of the amplitude of *ac* Josephson current ($\tilde{\mathcal{J}}$) versus $\frac{\omega}{2\Delta}$ and $\frac{\Omega}{2\Delta}$.

For $\frac{\Omega}{2\Delta} < 1$ the amplitude of the *ac* Josephson current diverges at points $\frac{\Omega+\omega}{2\Delta} = 1$, $\frac{\omega}{2\Delta} = 1$ and $\frac{\omega-\Omega}{2\Delta} = 1$. Also, as it is clearly observed, $\tilde{\mathcal{J}}$ changes its sign at two values of $\frac{\omega}{2\Delta}$ around one. However, for $\frac{\Omega}{2\Delta} > 1$ two divergencies emerge at points $\frac{\omega}{2\Delta} = 1$ and $\frac{\omega-\Omega}{2\Delta} = 1$. The amplitude vanishes and changes sign at two points around the $\frac{\omega-\Omega}{2\Delta} = 1$. These divergencies appear when the absorbed or emitted energy by the quasiparticles, due to interaction with the spin dynamics of SMM are close to the superconducting gap. At these situations, the resonance condition takes place in the junction and the current flowing through the junction diverges. So, tuning the values of the Ω and ω close to a resonance condition may leads to pumping a Josephson current trough the junction.

Adiabatic limit - In the adiabatic limit ($\frac{\Omega}{2\Delta} \ll 1$, $\frac{\omega}{2\Delta} \ll 1$), when the evolution in the system are very slow compared to the dwell time of the quasiparticles, there is no normal current flowing through the junction and the Josephson current is simplified as

$$i^J(t) = \pi\Delta[(T_0^2 - T_S^2) - T_{\perp}^2\frac{\Omega}{8\Delta} + T_{\perp}T_{\parallel}\frac{\Omega}{8\Delta}\vartheta\cos\omega t]\sin\chi. \quad (18)$$

Surprisingly, in the adiabatic limit the magnitudes of the *dc* and *ac* Josephson currents depend linearly on the precession frequency Ω and they are independent of the tilt angle oscillation frequency ω .

Conclusion - We introduced a charge current pump, a Josephson junction consisting of a single molecular magnet with spin nutation embedded between two spin singlet superconducting leads. The spin nutation, spin precession combined with an oscillation about it, is generated by applying a time dependent magnetic field to the single molecular magnet. The simultaneous effects of precession and oscillation cause to pump an *ac* normal and Josephson currents through the system. Varying the magnetic field enables us to control the magnitudes of the pumped currents. At resonance conditions some discontinuities and divergencies emerge in the normal and Josephson currents. Such behaviors appear due to the interaction of the quasiparticles with spin dynamics of the single molecular magnet and take place when the absorbed/emitted energy during the transferring is in the range of the superconducting gap.

The long relaxation times of the SMMs and small size of the junctions consisting of them make them favorable to use in molecular spintronics and quantum computing. Our introduced system have interesting properties and

would be important from practical point of view. The modulation of the Josephson current by applied magnetic field can make possible the single spin detection. Moreover, tuning the applied magnetic field to bring the system close to a resonance condition enable us to pump *dc* and *ac* normal and Josephson currents trough the system in a controllable way.

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