Pumping \textit{ac} Josephson current in the Single Molecular Magnets by spin nutation

B. Abdollahipour\textsuperscript{1*}, J. Abouie\textsuperscript{2,3*}, and A. A. Rostami\textsuperscript{4}

\textsuperscript{1}Department of Physics, University of Tabriz, Tabriz 51666-16471, Iran
\textsuperscript{2}Department of Physics, Institute for Advanced Studies in Basic Sciences (IASBS), Zanjan 45137-66731, Iran
\textsuperscript{3}School of Physics, Institute for Research in Fundamental Sciences (IPM), Tehran 19395-5531, Iran
\textsuperscript{4}Department of Physics, Shahrood University of Technology, Shahrood 36199-95161, Iran

We demonstrate that an \textit{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 \cite{1-4}. It has been introduced as a potential way to generate a dissipationless charge current in the nanoelectronic devices \cite{2}. 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 \cite{6}. In recent years, electron pumps consisting of different systems such as small semiconductor quantum dots \cite{7-9}, carbon nanotube quantum dot \cite{10}, one-dimensional interacting Lüttlinger liquid quantum wire \cite{11}, Josephson junctions with half-metallic ferromagnets \cite{12} and diffusive ferromagnets \cite{13}, and InAs nanowire embedded in a superconducting quantum interference device (SQUID) \cite{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 \cite{15} and classical \cite{16} and quantum information processing \cite{17}. The small sizes of these junctions are an advantage for their application. Compounds of single molecular magnet class \cite{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 \cite{20, 21}. The rich physics behind the magnetic behavior produces interesting effects such as negative differential conductance and complete current suppression \cite{22, 23}, which could be used in nanoelectronics.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig1}
\caption{(Color online) Color map of the amplitude of the \textit{ac} Josephson current $\bar{J}$ as a function of $\omega/\Delta$ and $\Omega/\Delta$. At $\frac{\Omega}{\Delta} \sim 1$ the amplitude goes down to very small values (infinity) for $\frac{\omega}{\Delta} \neq 1$. At $\frac{\omega}{\Delta} \sim 1$ the amplitude diverges for $\frac{\Omega}{\Delta} \neq 1$.}
\end{figure}

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 \textit{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

*These authors have contributed equally to this work.
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:

\[
\mathbf{S}(t) = S (\sin \theta(t) \cos \varphi(t), \sin \theta(t) \sin \varphi(t), \cos \theta(t)) . \tag{1}
\]

Where

\[
\varphi(t) = \Omega t, \quad \theta(t) = \theta_0 - \vartheta \cos \omega t,
\]

\[
\Omega = \gamma h_z \quad (\gamma \text{ denotes gyromagnetic ratio}) \text{ is the precession frequency around the } z \text{ axis and } \theta(t) \text{ is the time dependent tilt angle (angle between the spin and } z \text{ axis). The tilt angle oscillates about } \theta_0 \text{ with frequency } \omega \text{ and amplitude } \vartheta = \gamma h_0 / \omega. \quad \text{(See Fig. 2)} \quad \text{Indeed, this nutational motion of the spin is served as the pumping parameter in the system. Let us consider the following Hamiltonian for the Josephson junction:}

\[
H(t) = H_L + H_R + H_T(t) . \tag{3}
\]

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

\[
H_{\alpha} = \sum_{k,\sigma = \uparrow, \downarrow} \varepsilon_k c^\dagger_{\alpha,k,\sigma} c_{\alpha,k,\sigma} + \sum_k \left( \Delta^\dagger_{\alpha} c^\dagger_{\alpha,k,\uparrow} c_{\alpha,-k,\downarrow} + h.c. \right) \tag{4}
\]

where \(c^\dagger_{\alpha,k,\sigma}(c_{\alpha,k,\sigma})\) is the creation (annihilation) operator of an electron on the lead \(\alpha = L, R\) with momentum \(k\) and spin \(\sigma\). \(\varepsilon_k\) is the energy of single conduction electron and \(\Delta^\dagger_{\alpha} = \Delta e^{i\chi_{\alpha}}\) is the pair potential in which \(\chi_{\alpha}\) is the superconducting phase of the lead \(\alpha\). The two leads are weakly coupled via the tunneling Hamiltonian, \(H_T(t)\) which is given by

\[
H_T(t) = \sum_{k,k',\sigma,\sigma'} \left( c^\dagger_{R,k,\sigma} T_{\sigma\sigma'}(t) c_{L,k',\sigma'} + h.c. \right) \tag{5}
\]

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

\[
\hat{T}(t) = T_0 \mathbf{1} + T_S \mathbf{S}(t) \cdot \hat{\sigma}, \tag{6}
\]

where \(\mathbf{1}\) is the 2 \times 2 unit matrix, \(\hat{\sigma} = (\sigma_x, \sigma_y, \sigma_z)\) is the Pauli matrices and \(\mathbf{S}(t) = \frac{S}{|S|} \mathbf{S}\) 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 \(\alpha\) in two parts [20]

\[
I_\alpha(t) = I_\alpha^s(t) + I_\alpha^c(t) \tag{7}
\]

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

\[
A_\alpha(t) = \sum_{k,k',\sigma,\sigma'} \tau^\dagger_{\sigma,\sigma'}(t) T_{\sigma\sigma'}(t) c_{\alpha,k',\sigma'}(t), \tag{8}
\]

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

\[
U^{\alpha\sigma'}_{pp'}(t - t') = -i \Theta(t - t') \left\langle \left[ a_{\alpha,k,\sigma}(t), a_{p,p'}^{\dagger}(t') \right] \right\rangle , \tag{9}
\]
where, \( a_{\alpha,k}^{\sigma'}(t) = c_{\alpha,k}^{\dagger}(t) c_{\sigma,k',\sigma'}(t) \) and \( a_{p,p'}^{\sigma'}(t') = c_{\alpha,p}^{\dagger}(t') c_{\sigma',p',\sigma'}(t') \). Since there is no spin dependent interaction inside the superconducting leads, the retarded potential \( \bar{\Phi} \) is independent on spin indices and \( U_{\rho \rho'}^{\sigma}(t-t') = \delta_{\sigma \rho} \delta_{\rho \rho'} U_{\text{ret}}(t-t') \). The Fourier transformation of the Matsubara potential, with imaginary time, is defined as

\[
U(i\omega_n) = \frac{1}{\beta} \sum_{iq} g_R(k,iq-i\omega_n) g_L(k',iq) ,
\]

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

\[
i^s(t) = \Delta T_\perp \left[ 2T_{\perp} S\left( \frac{\Omega}{2\Delta} \right) + T_{||} S\left( \frac{\omega}{2\Delta} \right) \right] \cos \omega t ,
\]

where \( i^s = I^s/2\pi eN_L N_R \) as the density of states at the Fermi energy in the left (right) lead. To obtain this equation we have considered \( \theta/\theta_0 \ll 1 \). This approximations is fulfilled by the practical conditions of the system and do not make any restriction on it. The parameters \( T_{||} = T_S \cos \theta_0 \) 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 \( \theta_0 = n\pi \) \( (n = 0,1,\ldots) \). If the spin-flip transmission amplitude is nonzero \( (T_{\perp} \neq 0) \), depending on the strenth of precession frequency \( \Omega \) and tilt angle oscillation frequency \( \omega \), 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 \( S\left( \frac{\Omega}{2\Delta} \right) \) and \( S\left( \frac{\omega}{2\Delta} \right) \), which are given by

\[
S\left( \frac{\Omega}{2\Delta} \right) = 2S\left( \frac{\omega}{2\Delta} \right) - 2S\left( \frac{\Omega + \omega}{2\Delta} \right) - S\left( \frac{\Omega - \omega}{2\Delta} \right) \quad \text{and} \quad S\left( \frac{\omega}{2\Delta} \right) = \Theta(x-1) \left\{ s_{x+1} K(x) - (x+1) K(x) \right\} ,
\]

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 if \( \theta_0 = 0 \), the spin has only a precessing motion about the \( z \) axis without tilt angle oscillation. In this case the \( dc \) Josephson current \( i^J = 2\left[ (T_S^0 - T_S^\perp) - T_S^\perp \right] \) \( \sin \chi \) 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 \( \theta_0 = 0 \), depending on the values of \( \Omega \) and \( \omega \), different situations will appear depending on the values of \( \Omega \) and \( \omega \). 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 \( \Omega = 2\Delta \). 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 \( \Omega = 2\Delta \).

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

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

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

\[
\mathcal{X} (i\omega_n) = \frac{1}{\beta} \sum_{iq} \mathcal{F}_{\rho}^\dagger(k,iq) \mathcal{F}_{\rho'}(k',iq-i\omega_n) ,
\]

where \( \mathcal{F}_{\rho} \) and \( \mathcal{F}_{\rho'} \) are the anomalous Green’s functions in the leads \( \underline{26} \). The retarded potential is obtained by analytical continuation. Implementing the real part of the retarded potential

\[
\Re \left\{ \sum_{k,k'} X_{\text{ret}}(x) \right\} = \left\{ \begin{array}{ll} \pi N_L N_R \Delta K(x) & x < 1 \\ \pi N_L N_R \Delta \frac{1}{2} & x > 1 \end{array} \right.
\]

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

\[
i^J(t) = \left[ (T_S^0 - T_S^\perp) \Theta(0) - 2T_S^\perp \mathcal{J}\left( \frac{\Omega}{2\Delta} \right) \right] \cos \omega t + \mathcal{J}(0) \sin \chi
\]

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

\[
\mathcal{J}\left( \frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta} \right) = 2\mathcal{J}(0) - 2\mathcal{J}\left( \frac{\Omega}{2\Delta} \right) + 2\mathcal{J}\left( \frac{\omega}{2\Delta} \right) - \mathcal{J}\left( \frac{\Omega + \omega}{2\Delta} \right) - \mathcal{J}\left( \frac{\Omega - \omega}{2\Delta} \right).
\]

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 \( i^J = 2\left[ (T_S^0 - T_S^\perp) \Theta(0) - T_S^\perp \mathcal{J}\left( \frac{\Omega}{2\Delta} \right) \right] \) \( \sin \chi \) 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 \( \theta_0 \neq \frac{\pi}{2} \), depending on the values of \( \mathcal{J}\left( \frac{\Omega}{2\Delta}, \frac{\omega}{2\Delta} \right) \), 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 \( \bar{\mathcal{J}} \) vanishes for arbitrary values of \( \omega \) and
the Josephson current \( i^J = 2(T_0^2 - T_\parallel^2)\bar{\mathcal{J}}(0) \sin \chi \) is time-
independent. In Fig. (1), we have shown the density plot
of the amplitude of \( \text{ac Josephson current} (\bar{\mathcal{J}}) \) versus \( \frac{\omega}{2\Delta} \) and \( \frac{\Omega}{\omega} \).

For \( \frac{\omega}{2\Delta} < 1 \) the amplitude of the \( \text{ac Josephson current} \) diverges at points \( \frac{\omega}{2\Delta} = 1, \frac{\Omega}{\omega} = 1 \) and \( \frac{\omega - \Omega}{2\Delta} = 1 \). Also,
as it is clearly observed, \( \bar{\mathcal{J}} \) 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 \( \Omega \) and \( \omega \) close to a resonance condition may leads
to pumping a Josephson current trough the junction.

Adiabatic limit - In the adiabatic limit \( \frac{\Omega}{\omega} \ll 1, \frac{\omega}{2\Delta} \ll 1 \), 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

\[
\bar{i}^J(t) = \pi \Delta \left[ (T_0^2 - T_\parallel^2) - T_\perp \frac{\Omega}{2\Delta} + T_\parallel T_\perp \frac{\omega}{2\Delta} \cos \omega t \right] \sin \chi.
\]

(18)

Surprisingly, in the adiabatic limit the magnitudes of the \( \text{dc} \) and \( \text{ac Josephson currents} \) depend linearly on the pre-
cession frequency \( \Omega \) and they are independent of the tilt angle oscillation frequency \( \omega \).

Conclusion - We introduced a charge current pump, a
Josephson junction consisting of a single molecular mag-
net with spin nutation embedded between two spin sin-
glet superconducting leads. The spin nutation, spin pre-
cession combined with an oscillation about it, is gen-
erated by applying a time dependent magnetic field to
the single molecular magnet. The simultaneous effects of
precession and oscillation cause to pump an \( \text{ac} \) 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 dis-
continuities 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 ab-
sorbed/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 mag-
netic filed can make possible the single spin detection.
Moreover, tuning the applied magnetic filed to bring the
system close to a resonance condition enable us to pump
\( \text{dc} \) and \( \text{ac normal and Josephson currents} \) trough the sys-
tem in a controllable way.

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