Shapiro like steps reveals molecular nanomagnets’ spin dynamics

Babak Abdollahipour,1 a) Jahanfar Abouie,2 b) and Navid Ebrahimi2

1) Faculty of Physics, University of Tabriz, Tabriz 51666-16471, Iran
2) Department of Physics, Institute for Advanced Studies in Basic Sciences (IASBS), Zanjan 45137-66731, Iran

We present an accurate way to detect spin dynamics of a nutating molecular nanomagnet by inserting it in a tunnel Josephson junction and studying the current voltage (I-V) characteristic. The spin nutation of the molecular nanomagnet is generated by applying two circularly polarized magnetic fields. We demonstrate that modulation of the Josephson current by the nutation of the molecular nanomagnet’s spin appears as a stepwise structure like Shapiro steps in the I-V characteristic of the junction. Width and heights of these Shapiro-like steps are determined by two parameters of the spin nutation, frequency and amplitude of the nutation, which are simply tuned by the applied magnetic fields.

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Molecular nanomagnets have recently attracted intense attentions due to their large effective spin and long magnetization relaxation times which make them suitable for applications in quantum information processing and molecular spintronics.1,2 A crucial aspect of the research on molecular nanomagnets is determination of their spin dynamics. One of the possible methods to detect the spin dynamics of molecular nanomagnets is ferromagnetic-resonance experiment which is extensively used for thin ferromagnetic layers.3 Another powerful technique to investigate the spin dynamics in bulk samples is inelastic neutron scattering. In molecular nanomagnets, the dynamics are usually extrapolated by fitting inelastic neutron scattering spectra to a spin Hamiltonian and by performing calculations within the framework set by this model.2 Transport measurement through nanomagnets is an on-demand method to determine the magnetic state of a nanomagnet. An intrinsic limitation to the current measurement is associated to the high access resistance of the normal contacts. However, when the contacting leads become superconducting, long-range correlations can extend throughout the whole system by means of the proximity effect. This not only lifts the resistive limitation of normal-state contacts, but further paves the way to probe electron transport through a single molecule.2–6

Mutual interaction of the Josephson current flowing through the junctions and the molecular nanomagnet’s spin dynamics elicits several interesting phenomena such as the modulation of the Josephson current and generation of a circularly polarized ac spin current with the nanomagnet’s Larmor precession frequency.6,7 The effects of molecular nanomagnet’s spin nutation on the Josephson current flowing through the Josephson junction has been studied in Ref.8 It has been shown that the spin nutation of the molecular nanomagnet causes generation of an ac Josephson current through the junction, in addition to the dc one.

Irradiation of a microwave to a Josephson junction eventuates in Shapiro step structure in the current-voltage (I-V) characteristic. In thin ferromagnetic layer/superconductor Josephson junctions Shapiro steps reveal the magnetic response of the ferromagnet.7,9 They appear at voltages \( V = n(\hbar/2e)\Omega \) with the ferromagnetic resonance frequency \( \Omega \), integer \( n \), and the ratio of the Planck constant and the elementary charge, \( \hbar/e \). A small magnet in the proximity of a weak link with purely electromagnetic mutual interaction may be detected through Shapiro-like steps caused by the precession of the magnetic moment. The magnetic field of the nanomagnet alters the Josephson current flowing through the link, while the magnetic flux generated by the Josephson junction acts on the magnetic moment of the nanomagnet.10,11 The interplay between the ac Joseph-
son current and the magnetization precession of the nanomagnet shows a spin-polarized Shapiro steps and rich subgap structure in the I-V curve.\textsuperscript{21}

In this paper, we proffer a feasible way to detect directly the spin dynamics of a nanotunneling molecular nanomagnet by current measurement. By inserting the molecular nanomagnet in a tunnel Josephson junction, and investigating behavior of the current flowing through the junction we demonstrate that at low temperatures and in the tunneling limit the modulation of the Josephson current due to the nutation of the molecular nanomagnet, obviously seen as a stepwise structure in the I-V characteristic. We show that these Shapiro-like steps disappear when the molecular nanomagnet has only a precessing around z-axis.

**Spin nutation:** Spin nutation of the molecular nanomagnet is a result of its interaction with the following effective magnetic field:

$$\mathbf{h}_{\text{eff}}(t) = \mathbf{h}_z + \mathbf{h}_+(t) + \mathbf{h}_-(t),$$

where $\mathbf{h}_z$ is a static field along $z$ axis and includes the external magnetic field as well as all other contributions such as exchange interaction, crystal anisotropy, and magnetostatic interaction, and $\mathbf{h}_{\pm}(t) = -\frac{h_{xy}}{2} [\cos(\omega_{\pm} t) \mathbf{\hat{e}} + \sin(\omega_{\pm} t) \mathbf{\hat{g}}]$ are two circularly polarized fields with amplitude $\frac{h_{xy}}{2}$ and frequencies $\omega_\pm$. (see Fig. 1) Using the phenomenological Landau-Lifshitz equation, the spin dynamics of the molecular nanomagnet, in the absence of spin relaxation processes, is obtained as

$$\mathbf{S}(t) = \mathbf{S}(\sin \theta(t) \mathbf{cos } \Omega, \sin \theta(t) \mathbf{sin } \Omega, \mathbf{cos } \theta(t)),$$

where $\mathbf{S}$ is magnitude of the molecular nanomagnet’s spin, $\Omega = (\omega_+ - \omega_-)/2 = \gamma h_z$ is the precession frequency around z axis with gyromagnetic ratio $\gamma$, and $\theta(t)$ is the time-dependent tilt angle of which is given by $\theta(t) = \theta_0 - \delta \theta \cos \omega t$. Here, $\delta \theta = (\gamma h_{xy}/\omega)$ and $\omega = (\omega_+ + \omega_-)/2$ denote the amplitude and frequency of the nutation of the molecular nanomagnet’s spin around $\theta_0$, respectively.

**Josephson current:** Hamiltonian of the tunnel Josephson junction, schematically shown in Fig. 1, is given by:

$$H(t) = H_L + H_R + H_T(t),$$

where $H_L$ and $H_R$, are the BCS Hamiltonian of the left (L) and right (R) superconducting leads with identical amplitude of the pair potential $\Delta$ and phases $\chi_L$ and $\chi_R$. These Hamiltonians are written as:

$$H_{\alpha} = \sum_{k, \sigma = \uparrow, \downarrow} \varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{k} (\Delta_{\alpha} c^\dagger_{k\uparrow} c_{k\downarrow} + h.c.),$$

where $\varepsilon_k$ is the energy of a single conduction electron, and $c^\dagger_{k\sigma} (c_{k\sigma})$ is the creation (annihilation) operator of an electron in the lead $\alpha = \text{L, R}$ with momentum $k$ and spin $\sigma$. The two superconducting leads are weakly coupled via the tunneling Hamiltonian:

$$H_T(t) = \sum_{k, k', \sigma, \sigma'} (c^\dagger_{Rk\sigma} T_{\sigma\sigma'}(t) c_{Lk'\sigma'} + h.c.) ,$$

where $T_{\sigma\sigma'}(t)$ is a component of the time dependent tunneling matrix which transfers electrons through the system. The tunneling matrix can be written as:

$$\tilde{T}(t) = T_0 \mathbf{1} + T_S \mathbf{S}(t) \cdot \sigma,$$

where $\mathbf{1}$ is $2 \times 2$ unit matrix, $\mathbf{S}(t)$ is the unit vector along the molecular nanomagnet’s spin (Eq. 2), and $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ denotes vector of the Pauli’s spin operators. The parameter $T_0$ is spin independent transmission amplitude while $T_S$ denotes the amplitude of spin dependent transmission originating from exchange interaction between conduction electrons and the molecular nanomagnet’s spin.\textsuperscript{14}

In the tunneling limit the Josephson current reads:

$$I_\sigma(t) = -e \int_{-\infty}^{t} dt' \langle [A_\sigma(t), A_\sigma(t')] \rangle + h.c.),$$

where

$$A_\sigma(t) = \sum_{k, k', \sigma, \sigma'} c^\dagger_{k\sigma} (t) T_{\sigma\sigma'}(t) c_{k'\sigma'}(t).$$

In order to obtain an explicit form for $I_\sigma(t)$ we define the following retarded potential

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

where $a_{k\sigma}^{\rho}(t) = c^\dagger_{k\sigma}(t) c_{k'\sigma'}(t)$ and $\rho$ and $\rho'$ denote $\uparrow$ and $\downarrow$. This potential includes both triplet and singlet correlations. In the presence of a spin-active junction including spin-flip processes singlet correlations penetrating into the magnetic region convert to triplet correlations.\textsuperscript{22-24} Moreover, the interplay of the magnetization dynamics and transported carriers also results in the conversion of the spin-singlet to the spin-triplet correlations, which is accompanied by the absorption or emission of a magnon.\textsuperscript{25,26} These induced triplet correlations have the same magnitude as the singlet correlations at the interface, however they survive over a long range despite the singlet correlations. The triplet correlations also penetrate into the superconductors which have small amplitudes in comparison to the bulk singlet components.\textsuperscript{27}

As the junction considered here has two tunnel barriers, we can ignore the small effects of the triplet correlations induced in the superconducting leads. Thus, the retarded potential \textsuperscript{29} simplifies to $X_{\rho\sigma'}^{\sigma}(t-t') = \sigma \chi \delta_{\sigma, -\rho} \delta_{\sigma', -\rho'} X_{ret}(t-t')$, where $\sigma, \sigma' = \pm 1$. To the first order of the nutation amplitude ($\delta \theta$), the Josephson current is obtained as:

$$I_{J}(t) = I_c \sin \chi = (I_0 + I_1 \delta \theta \cos \omega t) \sin \chi ,$$
where $\chi (= \chi_R - \chi_L)$ is the phase difference between two superconducting leads. By defining $T_\parallel = T_S \cos \theta_0$ and $T_\perp = T_S \sin \theta_0$, the coefficients $I_0$ and $I_1$ are given by:

$$I_0 = 2e \left[ 2(T_0^2 - T_\parallel^2) \mathcal{R}(0) - 2T_\perp^2 \mathcal{R}(\hbar \Omega / 2\Delta) \right],$$

$$I_1 = -2eT_\perp T_\parallel \left[ 2(0) - 2(\mathcal{R}(\hbar \Omega / 2\Delta) + 2\mathcal{R}(\hbar \omega / 2\Delta)) \right],$$

$$-\mathcal{R}(\hbar \omega + \hbar \Omega / 2\Delta) - \mathcal{R}(\hbar \omega - \hbar \Omega / 2\Delta),$$

where $\mathcal{R}(x) = \Re \sum_{k,k'} X_{rel}(x)$ is the real part of the retarded potential which is obtained using the Matsubara Green’s functions and analytical continuation as:

$$\mathcal{R}(x) = \begin{cases} \pi N^2 \Delta K(x) & x < 1, \\ \pi N^2 \Delta K(\frac{1}{2}) & x > 1. \end{cases}$$

Here, $N$ is density of states at the Fermi energy in the left and right leads and $K(x)$ is the complete elliptic integral of the first kind.

In the absence of the circularly polarized magnetic fields, $\delta \theta$ is zero and the spin of the molecular nanomagnet has only a precession around $z$ axis. This eventuates in flowing of a $dc$ Josephson current through the junction. Applying the two circularly polarized magnetic fields $h_{\perp}(t)$, leads to the spin nutation of the molecular nanomagnet ($\delta \theta \neq 0$), and causes the generation of an $ac$ Josephson current through the junction, in addition to the $dc$ one. We interpret this effect as being due to evolution of Andreev bound states generated by the correlated Andreev reflections from the superconducting interfaces. Spin precession of the molecular nanomagnet affects the charge current by making transitions between the continuum states below the superconducting gap edge and the Andreev levels. When the molecular nanomagnet’s spin has a nutational motion, the Andreev bound states energies vary with the tilt angle oscillation which eventuate in the modulation of the Josephson current. 

**Phase dynamics:** In order to detect the spin dynamics of a nutating molecular nanomagnet we insert it in a tunnel Josephson junction and investigate the phase dynamics of the junction. The phase dynamics of a tunnel Josephson junction composed of two superconducting leads, having phase difference $\chi$, coupled through a tunnel barrier and subjected to a bias current $I$ is generally described by resistively shunted Josephson junction (RSJ) model. In RSJ model, a dissipative Josephson junction is modeled by a parallel circuit consisting of an ideal Josephson junction and a resistance $R_{\text{shunt}}$. The shunted resistance $R$ is indeed used to model leakage currents through the junction. By defining $I_c$ as the critical Josephson current, the phase dynamics of the Josephson junction is governed by the following equation:

$$I = I_c \sin \chi + \frac{\hbar}{2eR} \frac{d\chi}{dt},$$

where $I_c \sin \chi$ is Josephson current through an ideal Josephson junction and the second term represents the current passing through the resistance $V/R$, where we have used the Josephson relation $V = (\hbar/2e)d\chi/dt$. For a tunnel Josephson junction which has not driven externally, the critical Josephson current $I_c$ depends just on the junction parameters and it has no time dependence. For such a junction and in the case that $I > I_c$, the phase difference, given by $\chi = \arcsin(I/I_0)$, has not time dependence. Consequently, the voltage difference is zero. For $I > I_c$ the phase difference grows with time which results in a nonvanishing voltage difference. In this case the current-voltage relation takes the form $(V) = R \sqrt{T^2 - T_0^2}$ and it almost has a step at $V = 0$. If the junction is driven by an $ac$ current such that $(I) = I_0 + I_1 \cos(\omega t)$, here $I_0$ is a static current and $I_1$ is the amplitude of the oscillation around it, the I-V characteristic of the junction show an stepwise structure called Shapiro step. These steps occur precisely when the average voltage match $(V) = n(\hbar \omega / 2e)$, where $n$ stands for integer values.

Making use of the Josephson current given by Eq. (10) the phase dynamics of the tunnel Josephson junction driven by the external magnetic fields (Eq. (11)) is given by the following rescaled equation:

$$i = (1 + i_1 \cos ft') \sin \chi + \frac{d\chi}{dt'},$$

where $i(= I/I_0)$ is the dimensionless bias current, $i_1 = \delta \theta I_1/I_0$, $f = \omega / \alpha$, $t' = ct$, and $\alpha = 2eR_{\text{shunt}}/\hbar$. Solving Eq. (14) numerically the rescaled voltages $v(= V/R_{\text{shunt}})$ are obtained by time averaging on $d\chi/dt'$, for different bias currents $i$. We have plotted in Fig. 2 the I-V characteristic for different values of $i_1$ and $f$. For easier comparison we have juxtaposed the frames of $f \sim 0$, $f = 0.5$ and $f = 1.5$. For $i_1 \neq 0$ and $f \neq 0$, as a result of the molecular nanomagnet’s spin nutation, Shapiro-like steps
appear at voltages $v = nf$, where $n$ is an integer number (see Fig. 2(b),(c)). The width and heights of these steps are directly controlled by two parameters $\delta v = f$ and $\delta i \propto i_1$. These Shapiro-like steps disappear when $\omega$ or $\delta \theta$ is zero, i.e. when the molecular nanomagnet has only a precession around $z$ axis. Indeed the spin nutation of the molecular nanomagnet is essential for the emergence of the Shapiro-like steps on the I-V characteristic. To become more clear we glance at the time dependence of the phase difference. As a result of the oscillatory term of the Josephson current, the current and voltage has been rescaled in our results the current and voltage is given by:

$$\chi(t) = \chi_0 + vt' + \delta \chi \sin ft'$$

where $\chi_0$ is a constant, $v$ is the rescaled time-averaged voltage and $\delta \chi$ is the amplitude of the phase modulation. Inserting this expression in Eq. (14) the time dependent part of the Josephson current, $i_1 \cos ft' \sin \chi$, can be expanded as

$$i_1 e^{\pm i\chi \pm i ft'} = i_1 \sum_{n=-\infty}^{\infty} J_n(\delta \chi) e^{i(\pm \chi_0 \pm vt' \mp n ft')},$$

where $J_n(x)$ is an ordinary Bessel function. The dc component of the Josephson current is obtained by time averaging on the right hand side of Eq. (16). The time average is non-zero at voltages $v = nf$ ($V = n \frac{h}{2e}$) where the Shapiro-like steps appear on the I-V curve.

According to ferromagnetic-resonance experiments (for example see Ref. [5]), the typical values of the precession frequencies of a ferromagnetic thin film in the proximity of a superconductor are in the range of tens of GHz which corresponds to a magnetic field of about 100 Gauss. Therefore the order of magnitude of the molecular nanomagnet’s precession and nutation energies, $\hbar \Omega$ and $\hbar \omega$, are about $\sim 10^{-6}$eV which result in Shapiro-like steps with width of $\sim 1 \mu$V. The heights of the Shapiro-like steps are proportional to the nutation amplitude $\delta \theta$, which can be adjusted by the amplitude of the circularly polarized magnetic fields ($\delta \theta = \gamma h_{xy}/\omega$). In our results the current and voltage has been rescaled by the dc josephson current $I_0$ and $R I_0$, respectively. The typical values of $I_0$ and $R$ are in the range of $\sim nA$ and 1 Ohm which result in the first step hight to be of the order of $1nA$. The order of magnitude of the width and heights of Shapiro-like steps are in ranges that are simply accessible in the experiment, thereby the realization of the effect presented above is feasible.

Now let us give a discussion on the possibility of the molecular nanomagnet-superconductor Josephson junction fabrication. In spite of several transport measurements on molecular nanomagnets connected to normal leads the only practical transport measurements on the molecular nanomagnets coupled to the superconducting leads has been performed for the magnetic $C_{60}$ fullerene injected in the electromigrated gold break junctions. Superconductivity is typically induced in these junctions by means of the proximity effect and molecular nanomagnets are coupled to the gold nanowires. Very recently it has been demonstrated that electronic transport through molecular nanomagnets can be extended to superconducting electrodes by combining gold with molybdenum-rhenium (MoRe). This combination induces proximity-effect superconductivity in the gold to temperatures of at least 4.6K and magnetic fields of 6T, improving on previously reported aluminum based superconducting nanojunctions.

Recently, it has been shown that a spin current is generated in the superconducting leads by spin precession of the molecular nanomagnet. The flowing spin current induces a torque on the nanomagnet and changes its time evolution. This back-action effect alters the parameters of the spin dynamics ($\Omega$ and $\omega$). It should be noted that although the back-action affects the motion of the nanomagnet’s spin, it does not disturb the spin nutational motion. Therefore, we expect that the Shapiro-like steps appear at voltages slightly different from $V = n \frac{h}{2e}$. The interaction of the spin with its environment such as exchange field and magnetic anisotropy can be considered by introducing a Gilbert damping constant. In this case, the spin dynamics of the molecular nanomagnet is given by the Landau-Lifshitz-Gilbert equation. Presence of the damping will change the amplitude of the nutation, but not its frequency. Thus, the interactions alters the heights of the steps and not their width, thereby they can not disturb the observed stepwise structure.

**Conclusion:** In conclusion, we have presented a feasible way to directly detect the spin dynamics of a mutating molecular nanomagnet. By fabricating a molecular nanomagnet-superconductor Josephson junction and measuring the voltage difference and Josephson current through the junction, we are able to obtain the current-voltage characteristic of the junction. The interplay of the Josephson current and the spin nutation shows itself as a stepwise structure on the I-V characteristic, in which the width and heights of the steps give directly the precession and nutation frequencies of the molecular nanomagnet’s spin. Our results would be of quite important for experimentalists and provide a direct way to detect the spin dynamics of a single molecular nanomagnet.

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