Spin backflow: a non-Markovian effect on spin pumping

Kazunari Hashimoto,1 Gen Tatara,2 and Chikako Uchiyama1,3

1Graduate School of Interdisciplinary Research, University of Yamanashi, Kofu 400-8511, Japan
2RIKEN Center for Emerging Matter Science (CEMS), 2-1 Hirosawa, Wako 351-0198, Japan
3National Institute of Informatics, 2-1-2 Hitotsubashi, Chiyoda-ku, Tokyo 101-8430, Japan

(Dated: January 16, 2019)

The miniaturization of spintronic devices, specifically, nanoscale devices employing spintronics, has attracted intensive attention from a scientific as well as engineering perspective. In this paper, we study non-Markovian effect on spin pumping to describe spin current generation driven by arbitrary precession frequency of magnetization in a quantum dot attached to an electron lead. Although the Markovian approximation can be used when driving is sufficiently slow compared with relaxation times in electron tunneling, recent developments in nano-spintronic devices show that we need to include non-Markovian effects. In contrast to the one-way-only nature of the spin current generation under the Markovian dynamics, we find that the non-Markovian dynamics exhibits a temporal backflow of spin, call spin backflow for brevity. We capture the phenomenon by introducing its quantifier, and show that the backflow significantly reduces the amount of spin current when the frequency exceeds the relaxation rate. This prevents unphysical divergence of the spin current in the high frequency limit that occurs under the Markovian approximation. We believe our analysis provides an understanding of the spin pumping particularly in regard to producing a more efficient spin current generation over shorter time scales by going beyond the conventional Markovian approximation.

I. INTRODUCTION

Controlling the electron transport in nano-systems represents a promising advance for future electronics. Its major application is the single-electron transistor,1,2 which would enable extreme downsizing and ultra-low-power consumption of computing devices. An ambitious research field with this direction in mind seeks to incorporate magnetic components into nano-electronic devices.3,4 It aims to boost the conventional nano-electronics devices by exploiting the spin degrees of freedom in addition to the electronic charge. Such devices have been implemented using ferromagnetic electrodes attached to non-magnetic islands,5 ferromagnetic quantum dots,6,7 and single-molecule magnets.8

The generation of spin current is the important aspect in nano-spintronics. To date, numerous efforts have been made to realize spin pumping in nano-systems9–25 based on the typical spin-pumping protocol of bulk systems with the precession of the magnetization of a ferromagnet attached to a normal metal lead.26,27 In contributing to these attempts, we have focused on a minimum model describing spin pumping in a nano-system consisting of an electron lead attached to a two-level system (quantum dot) subjected to a rotating magnetic field.28–30

In conventional studies on the minimum model, spin pumping has been formulated using the adiabatic approximation, which requires the rotation frequency of the magnetic field \( \Omega \) to be small compared with the characteristic energy scale \( \delta E \) over which the stationary scattering property of an electron by the quantum dot changes significantly, i.e., \( \Omega \ll \delta E / \hbar \). Underlying this condition is an implicit assumption, specifically, the relaxation time \( \tau_r \) of the electron distribution in the dot by tunneling to the lead is infinitely slow compared with the rotation, \( \tau_r^{-1} \ll \Omega \). Because it is impossible to set the relaxation time to infinity, we studied the effect of its finiteness in Ref.29 by evaluating the non-adiabatic effect up to \( \Omega \lesssim \tau_r^{-1} \) formulated subject to the Born–Markov approximation; see for example.30 In consequence, we showed that spin pumping is an entirely non-adiabatic effect and the spin current exhibits an oscillatory dependence on \( \Omega \), thereby indicating an enhancement in the spin current.

In the present study, we examined the non-Markovian effect on spin pumping by removing the Markovian approximation from the formulation. The Markovian approximation is only valid when the time scale of the relevant dynamics is sufficiently longer than the relaxation time of the dot as well as the correlation time of the lead. Therefore, breakdown occurs either for a rapidly rotating magnetic field when the relaxation time is exceeded or for a structured lead with a rather long correlation time, which often occurs for nano-spintronics systems. Indeed, in a single molecule magnet, the relaxation rate (\( \gamma_r \approx 0.36 \times 10^9 \text{ s}^{-1} \)) is much smaller than the typical frequency of microwaves (\( \nu \approx 10 \text{ GHz} \)) that may excite precessions in the magnet’s magnetic core.31

Among several non-Markovian effects,32–44, we focus on the effects revealed as backflow,45,46 which reflects the partial reversibility allowing back-and-forth transfer of information and energy unlike the one-way-only transfers of electrons under Markovian dynamics. Although these studies treat undriven systems, the backflow may significantly affect electron transport in a constantly driven system because non-Markovian effects dominate the initial stage of the relaxation process following a given external disturbance. The question arises: what is the role of the backflow in a constantly driven system such as in spin pumping? To answer the ques-
FIG. 1. Schematic drawing of the minimum model. The model consists of a ferromagnetic quantum dot attached to an electron lead. The dot has a dynamic magnetization \( M(t) \) that rotates around the \( z \)-axis with a period \( T \). The number of transferred electrons with spin magnetic moment \( \uparrow (\downarrow) \) is captured by the counting field (see Formalism).

We formulate a non-Markovian dynamics of electron transfer in the minimum model by using the full counting statistics\(^5\), which provides short time behavior description of partial reversibility allowing spin transfer back from lead to dot which we call spin backflow. We find that the non-Markovian dynamics enables a physically reasonable description of spin pumping for all frequency range.

II. MODEL

We consider a simple model of spin pumping (Fig. 1) that describes a quantum dot with a dynamic magnetization attached to an electron lead\(^6,7\). In the quantum dot, the electron is spin polarized because of the d-s exchange interaction with the magnetization and is represented by a two-component creation and annihilation operators \( d^\dagger = (d_\uparrow ^\dagger, d_\downarrow ^\dagger) \) and \( d \), where \( \uparrow \) or \( \downarrow \) represents the direction of the spin magnetic moment of the electron parallel or antiparallel to the \( z \)-axis.

The Hamiltonian \( H(t) = H_d(t) + H_1 + H_t \) comprises three terms: \( H_d(t) \) describing the dot is defined by \( H_d(t) = d^\dagger (c_d - M(t) \cdot \sigma) d \), where \( c_d \) is the unpolarized energy of a dot electron, \( M(t) \equiv M(\sin \theta \cos \phi(t), \sin \theta \sin \phi(t), \cos \theta) \), and \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the vector of Pauli matrices. The electron lead is described by the term \( H_1 = \sum_{\sigma=\uparrow,\downarrow} \sum_k \epsilon_k c_{\sigma,k}^\dagger c_{\sigma,k} \), where \( c_{\sigma,k}^\dagger \) and \( c_{\sigma,k} \) with \( \sigma = \uparrow \) or \( \downarrow \) represents the creation and annihilation operators of a lead electron with energy \( \epsilon_k \). The dot–lead interaction is assumed to be spin conserving with \( H_t = \sum_{\sigma} \sum_k \hbar v_k (d_k^\dagger c_{\uparrow,k}^\dagger + c_{\sigma,k}^\dagger d_\downarrow) \), where \( \hbar v_k \) is the coupling strength, which we assume to be weak.

III. FORMALISM

We formulate the spin pumping in terms of the full counting statistics\(^8\) that is to provide the statistics for the net number of electrons transferred between the dot and the lead (Fig. 1) within a certain time interval. It uses the outcomes of two successive projective measurements of the number of electrons in the lead. The measurement scheme is as follows. At \( t = t_i \), we perform a measurement of the number of electrons with spin \( \sigma \), represented by operator \( N_\sigma \equiv \sum_k c_{\sigma,k}^\dagger c_{\sigma,k} \), to obtain an outcome \( n_{\sigma,i} \). Between \( t_i \leq t \leq t_{i+1} \), the system undergoes a unitary time evolution subject to the interaction between dot and lead. At \( t = t_{i+1} \), we perform another measurement of \( N_\sigma \) to obtain another outcome \( n_{\sigma,i+1} \). The net number of transferred electrons during \( \delta t = t_{i+1} - t_i \) is therefore given by \( \Delta n_{\sigma,i} \equiv n_{\sigma,i+1} - n_{\sigma,i} \), the sign of which is chosen to be positive when an electron is transferred from dot to lead. The cumulants of \( \Delta n_{\sigma,i} \) are provided by its cumulant generating function \( \mathcal{S}(\lambda_\sigma) \equiv \ln \int_\mathcal{Z} P(\Delta n_{\sigma,i}) e^{i\lambda_\sigma \Delta n_{\sigma,i}} d\Delta n_{\sigma,i} \), where \( P(\Delta n_{\sigma,i}) \) is the probability distribution function of \( \Delta n_{\sigma,i} \) and \( \lambda_\sigma \) is the counting field associated with \( N_\sigma \), which gives the mean value of \( \Delta n_{\sigma,i} \) by its first derivative as \( \langle \Delta n_{\sigma,i} \rangle = \partial \mathcal{S}(\lambda_\sigma)/\partial (i\lambda_\sigma)|_{\lambda_\sigma=0} \).

In the formalism, the cumulant generating function is rewritten as \( \mathcal{S}(\lambda_\sigma) = \ln \text{Tr}_d \rho(t_{i+1}) \), where \( \text{Tr}_d \) denotes the trace operation over the states of the dot, and operator \( \rho(\lambda_\sigma)(t) \) is a solution of the quantum master equation of time–convolutionless type\(^9,10\) modified to include the counting field, \( \partial \rho(\lambda_\sigma)(t)/\partial t = \xi(\lambda_\sigma)(t)\rho(\lambda_\sigma)(t)\delta(t,\lambda_\sigma) \), for a factorized initial condition between dot and lead at \( t_i \) as \( \rho^{(0)}(t_i) \equiv \rho_0 \), with \( \rho_0 \equiv \text{exp}(-\beta H_d)/\text{Tr}[\text{exp}(-\beta H_d)] \). By taking cumulants of the interaction of up to second order, the generator of the time evolution of \( \rho(\lambda_\sigma)(t) \) is given by \( \xi(\lambda_\sigma)(t) \equiv -i\hbar^{-1}[H_d, \rho] + K_2(\lambda_\sigma)(t)\rho \), where \( K_2(\lambda_\sigma)(t) \equiv -\hbar^{-2} \int_0^t dt_1 \text{Tr}_d[H_1, [H_1, (t_1], \rho \otimes \rho_0^{eq}]_{\lambda_\sigma}]_\sigma \) is the memory kernel with definitions \( H_1(t) \equiv e^{i(\lambda_\sigma - 1)iH_d t} H_1 e^{-i(\lambda_\sigma - 1)iH_d t} \), \( A_\sigma^{eq} \equiv A^{(\lambda_\sigma)} B - B A^{(\lambda_\sigma)} \), and \( A^{(\lambda_\sigma)} \equiv e^{i\lambda_\sigma N_\sigma}/2 A - e^{-i\lambda_\sigma N_\sigma}/2 B \). The memory kernel reflects the finiteness of the correlation time of the dot–lead interaction. The above formalism gives the mean value of \( \Delta n_{\sigma,i} \) as \( \langle \Delta n_{\sigma,i} \rangle = \int_{t_i}^{t_{i+1}} J_\sigma(s) ds \) with a temporal flow of electrons with spin \( \sigma \), \( J_\sigma(t) \equiv \text{Tr}_d \{ \partial \rho(\lambda_\sigma)(t)/\partial (i\lambda_\sigma)\}_{\lambda_\sigma=0} \rho^{(0)}(t) \), the sign of which is chosen to be positive when electrons are transferred from dot to lead\(^11\).

To formulate spin pumping based on the above framework, we consider a step–like change in the direction of \( M(t) \) around the \( z \)-axis; specifically, dividing the period \( T \) into \( N \) intervals, \( t_1 \leq t \leq t_{N+1} (i = 1, 2, \cdots, N) \) with \( t_1 = 0 \) and \( t_{N+1} = T \), fixing the direction of \( M(t) \) during each interval, and changing \( \phi \) at each \( t_i \) discretely with substitution \( \phi_i = \phi_{i-1} + \delta \phi \) with \( \phi_0 = 0 \), \( \phi_N = 2\pi \), and \( \delta \phi \equiv 2\pi/N \). The time dependence of \( \phi(t) \) is expressed as \( \phi(t) = (1 + [t/\delta t]) \phi_0 \), where \( |x| \equiv \max\{n \in \mathbb{Z} | n \leq x\} \), and \( \delta t = T/N \). Assuming the factorized form of the total density matrix at each \( t = t_i \), we obtain the mean number \( \langle \Delta n_{\sigma,i} \rangle \). Its summation over a single cycle \( \langle \Delta n_{\sigma} \rangle \equiv \sum_{i=1}^{N} \langle \Delta n_{\sigma,i} \rangle \) gives the total number of electrons with spin \( \sigma \) transferred during the cycle. We therefore define the generated spin current by

\[
I_\sigma \equiv \frac{\langle \Delta n_{\sigma} \rangle - \langle \Delta n_{\sigma} \rangle}{T},
\]
which enables us to introduce a temporal flow of spin by
its time derivative, specifically,
\[
J_{\text{spin}}(t) \equiv \tau \frac{dI_{\uparrow}}{dt} = J_{\uparrow}(t) - J_{\downarrow}(t).
\]  

IV. SPIN BACKFLOW

Here we introduce the concept of spin backflow, the concept of which is different from the spin-current backflow introduced in Ref. 2, see Discussions. As shown above, the memory kernel, \( K_2^{(\lambda_0)}(t) \), in our formalism includes the finite correlation time of the dot–lead interaction. This enables us to describe the time interval in which partial reversibility remains, corresponding to the exchange of an electron between dot and lead—the so-called non-Markovian dynamics. Partial reversibility, allowing the back and forth transfer of an electron, is revealed with the sign reversal of \( K_2^{(\lambda_0)}(t) \), which turns out to be the dynamical change in direction of the flow of spin. We call this return of the electron from the lead a spin backflow. This feature is quite different from the conventional notion of the one-way-only transfer of an electron under the Markovian approximation, for which the time-dependence of \( K^{(\lambda_0)}(t) \) is removed by taking the long-time limit of the memory kernel, specifically, \( \lim_{t \to \infty} K^{(\lambda_0)}(t) \).

The spin backflow is captured by monitoring the sign change of the temporal flow of spin \( J_{\text{spin}}(t) \), Eq. \( \text{(2)} \). When the flow is positive, spin is transferred from dot to lead; conversely, when the flow is negative, spin is transferred from lead to dot. In contrast, under the Markovian approximation, we expect that the sign of the temporal flow remains the same during its time evolution.

V. NUMERICAL RESULTS

Let us now analyze spin backflow by numerically evaluating the flow of spin \( J_{\text{spin}}(t) \) and how it affects the frequency dependence of the spin current \( I_{\uparrow} \), Eq. \( \text{(1)} \). In each instance, we also present numerical results obtained subject to the Markovian approximation as a reference for comparison with the non-Markovian result.

To describe the dot–lead coupling, we use the Ohmic spectral density with an exponential cutoff \( v(\omega) \equiv \sum_k v_k^2 \delta(\omega - \omega_k) = \lambda \omega \exp[-\omega/\omega_c] \), where \( \lambda \) is the coupling strength and \( \omega_c \) is the cutoff frequency. For the numerical calculation, we choose 2M, the energy difference between the spin-\( \uparrow \) and \( \downarrow \) states in the dot, as an energy unit, and we identify the parameters normalized by the unit with an overbar (see note 22). Specific values of the parameters are given in the figure captions. As an initial condition, we set the dot in a steady state 22.

As we are focusing on the spin transfer driven by the rotating magnetization, setting the dot in a steady state 22 excludes any transient spin transfer caused by the dot–lead contact at \( \bar{t} = 0 \). Under the initial condition, the net charge transfer \( \langle \Delta n_{\uparrow} \rangle + \langle \Delta n_{\downarrow} \rangle \) is zero because the charge is conserved in the lead. Nevertheless, a spin current \( I_{\uparrow} \) is generated because equal amounts of spin-\( \uparrow \) and spin-\( \downarrow \) electrons are transferred in the opposite directions, i.e., \( \langle \Delta n_{\uparrow} \rangle = -\langle \Delta n_{\downarrow} \rangle \) because spin flips in the dot are driven by the rotating magnetization.

Let us first examine the temporal flow of spin \( J_{\text{spin}}(t) \), Eq. \( \text{(2)} \). We plot its time evolution under the non-Markovian analysis [Fig. 2(a)] as well as corresponding Markovian analysis [Fig. 2(b)]. Both time evolutions are given for a single time interval for the step-like rotation of the magnetization, setting this interval to be larger than the relaxation time (specifically, \( \tau_r \approx 20 \) and \( \delta \bar{t} = 40 \)).

Fig. 2(a) exhibits two different oscillations: the larger oscillation with the longer period reflects the back and forth transfer of spin caused by the non-Markovian dynamics arising from the dot–lead coupling, whereas the smaller oscillation with the shorter period reflects the periodic transition between the spin-\( \uparrow \) and spin-\( \downarrow \) states in the dot under a magnetization with Larmor frequency \( 2M/\hbar \). In contrast, Fig. 2(b) only exhibits the Larmor precession.

Fig. 2(a) also shows that the flow of spin under non-Markovian dynamics starts from zero at \( \bar{t} = 0 \), which properly reflects the moment when the dynamics starts from the steady state. The gray-colored region identifies negative flow of spin, \( J_{\text{spin}}(t) < 0 \), which we call spin backflow, where the spin current flows back from the lead to the dot.

In contrast, regarding the Markovian dynamics [Fig. 2(b)], we find that the spin starts flowing with a finite impetus at \( \bar{t} = 0 \), always taking positive values during its time evolution. Physically, this indicates that the spin is always transferred from dot to lead without backflow.

Focusing on the initial short-time behavior, direction of the flow of spin in the non-Markovian dynamics (\( J_{\text{spin}} < 0 \)) is opposite to that in the Markovian dynamics (\( J_{\text{spin}} > 0 \)).
The red and blue dashed lines mark the non-Markovian and Markovian results, respectively. Inset (i) presents a magnification of the interval $0 \leq \bar{\Omega} \leq 0.005$; inset (ii) presents the frequency dependence up to higher frequencies. Both results exhibit oscillations that depend on $\bar{\Omega}$ for $\bar{\Omega} \gtrsim 0.002$ and is a consequence of Rabi oscillations in the dot. The results coincide in the linear regime ($\bar{\Omega} \lesssim 0.002$) whereas they deviate in the oscillating regime. The parameter values are the same as in Fig. 2.

We next discuss how the spin backflow affects the spin current generation. For this purpose, we evaluated the frequency dependence of the spin current $I_\uparrow$, Eq. (1), in Fig. 3, where the red solid (blue dashed) line presents the spin current under the non-Markovian (Markovian) dynamics, respectively. Inset (i) is a magnification of the lower-frequency regime and inset (ii) shows the frequency dependence up to higher frequencies.

For both dynamics, we find a common qualitative feature, i.e., the linear dependence on $\bar{\Omega}$ gradually changes to an oscillatory dependence for higher frequencies (around $\bar{\Omega} \gtrsim 0.002$ in Fig. 3), which is explained as a consequence of the relaxation time of the population in the dot, $\bar{\tau}_r$. For lower frequencies, for which the time interval is sufficiently larger than the relaxation time as $\delta t \gg \bar{\tau}_r$, the numerator of Eq. (1) becomes constant because the flow of spin $J_{\text{spin}}(t)$ has already vanished at a certain $t < \delta t$ (see Fig. 2), which results in the linear dependence of $I_\uparrow$ on $\bar{\Omega}$. As $\bar{\Omega}$ becomes larger and the time interval satisfies $\delta t \lesssim \bar{\tau}_r$, the angle of magnetization $\phi$ changes during relaxation. In this situation, we have two extreme features; when $\delta t$ is an integer multiple of the period of a spin flip $\hbar/2M$, we have resonance enhancement of the spin flip by changing $\phi$ to exhibit a maximum, whereas it is anti-resonantly suppressed to display a minimum when $\delta t$ is a half-integer multiple of the period\textsuperscript{35}.

Comparing the non-Markovian and Markovian analyses, we find a coincidence in the lower frequency (linear) regime, whereas over the higher frequency regime the Markovian and non-Markovian analyses deviate, the deviation being quite significant in inset (ii). The Markovian analysis diverges with respect to $\Omega$, whereas the non-Markovian analysis is totally suppressed. We explain the physical origin of this divergence by the non-zero flow of spin at $\bar{t} = 0$ for Markovian dynamics and by the suppression in the spin backflow for non-Markovian dynamics.

We note the following three points: (A) The amount of spin backflow monotonically increases as the cutoff frequency $\omega_c$ increases and, it saturates towards a certain value in the limit $\omega_c/\omega_c \rightarrow 0$. This is because the dot–lead coupling represented by $\nu((\pm M)/\hbar)$ increases towards the Ohmic (linear) function $\lambda((\pm M)/\hbar)$ in the limit. (B) The spin current decreases as the lead temperature increases because the numeric difference between the transferred electrons with spin-up and spin-down approaches zero, for the reason that the two electron populations in the lead during the interaction windows of the spin-up and spin-down electrons move closer. (C) The spin polarization of the current exhibits a $\theta$ dependence in that for $0 < \theta < \pi/2$ the spin polarization is antiparallel to the $z$-axis, whereas for $\pi/2 < \theta < \pi$ the spin polarization is parallel to the $z$-axis for the non-Markovian analysis, as shown in\textsuperscript{27} for the Markovian analysis.

VI. DISCUSSIONS

In the context of spin pumping in bulk systems, some researchers have studied the “backflow (or backscatter) of the spin current” because of the finite size of the electron reservoir and the slow modulation for the system to follow the precession sufficiently\textsuperscript{8,23}. They have argued that, when the pumped angular momentum does not quickly dissipate to the lead, a nonvanishing spin accumulation may build up in the lead. For a sufficiently slow precession, the spin imbalance through spin accumulation may flow back into the ferromagnet, canceling the generated spin current as the system is always in the steady state. The behavior of this backflow in spin current is different from the spin backflow studied in this work in regard to two points: (i) the latter occurs even for an ideal reservoir in which the pumped spin is absorbed entirely, whereas the former is caused by the accumulation of spin angular momentum in the finite reservoir, and (ii) the latter becomes significant for rapid precession, whereas the former requires a sufficiently slow precession. Therefore, the spin backflow studied in this paper is a completely independent concept from the conventional backflow of spin current. When one considers a non-ideal reservoir of finite size and a moderately rapid precession, both backflow processes may coexist. A study of the situation is left for a future investigation.

Although we have focused on the spin backflow in this work, the concept of backflow itself is a universal feature of quantum transport under the non-Markovian dynamics. Indeed, some researchers have studied the backflow of information\textsuperscript{34} and energy\textsuperscript{44} in undriven systems. Because this is the first study of backflow in a driven system, we conjecture that our main result, the reduction of the
pumped quantity because of backflow, holds for a wide range of driven systems. We will discuss the universality of our results elsewhere.

The steplike rotation reduces to a continuous rotation in a limit $\delta t \to 0$, $\delta \phi \to 0$ with $T = \text{constant}$. With a non-zero Markovian flow at $t = 0$, the limit leads to a divergence of the spin current under the Markovian approximation (Fig. 3). To avoid this divergence, we need to include the non-Markovian effect.

VII. CONCLUSIONS

Focusing on spin backflow, we have examined the role of the non-Markovian effect on the spin pumping under a precessing magnetization. In evaluating the frequency dependence of the pumped spin current, we compared the results obtained from our non-Markovian analysis with those under a corresponding Markovian analysis. Our numerical result shows that spin backflow does not contribute to the net amount of spin current in the low-frequency regime where $\delta t \gtrsim \tau_r$, whereas it significantly reduces the spin current in the high-frequency regime where $\delta t \lesssim \tau_r$. This provides the physically reasonable description of the spin pumping over all frequencies, which could not be achieved by conventional Markovian approximation.

ACKNOWLEDGMENTS

This work was supported by the Grant-in-Aid for Challenging Exploratory Research (No. 16K13853) and partially supported by the Grant-in-Aid for Scientific Research on Innovative Areas Science of Hybrid Quantum Systems (No. 18H04290).

1. T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. 59, 109 (1987).
2. K. J. Dempsey, D. Ciudad, and C. H. Marrows, Phil. Trans. R. Soc. A 369, 3150 (2011).
3. D. D. Awschalom, L. C. Bassett, A. S. Dzurak, E. L. Hu, J. R. Petta, Science 339, 1174 (2013).
4. C. D. Chen, W. Kuo, D. S. Chung, J. H. Shyu, and C. S. Wu, Phys. Rev. Lett. 81, 047004 (2002).
5. H. Yang, S. -H. Yang, and S. S. Parkin, Nano Lett. 8, 340 (2008).
6. L. Bogani and W. Wernsdorfer, Nat. Mater. 7, 179 (2008).
7. Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, Phys. Rev. Lett. 88, 117601 (2002).
8. Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, Phys. Rev. B 66, 224403 (2002).
9. S. Maekawa, H. Adachi, K. Uchida, J. Ieda, and E. Saitoh, J. Soc. Phys. Jpn. 82, 102002 (2013).
10. E. R. Mucciolo, C. Chamont, and C. M. Marcus, Phys. Rev. Lett. 89, 146802 (2002).
11. B. Wang, J. Wang, and H. Guo, Phys. Rev. B 67, 092408 (2003).
12. P. Zhang, Q. K. Xue, and X. C. Xie, Phys. Rev. Lett. 91, 196602 (2003).
13. E. Cota, R. Aguado, and G. Platero, Phys. Rev. Lett. 94, 107202 (2005).
14. J. Splettstoesser, M. Governale, and J. König, Phys. Rev. B 77, 195320 (2008).
15. M. Braun and G. Murkard, Phys. Rev. Lett. 101, 036802 (2008).
16. K. Hattori, Phys. Rev. B 78, 155321 (2008).
17. R. Riwar and J. Splettstoesser, Phys. Rev. B 82, 205308 (2010).
18. J. Fransson and M. Galperin, Phys. Rev. B 81, 075311 (2010).
19. N. Winkler, M. Governale, and J. König, Phys. Rev. B 87, 155428 (2013).
20. S. Rojek, M. Governale, and J. König, Phys. Status Solidi B 251, 1912 (2013).
21. S. Rojek, J. König, and A. Shnirman, Phys. Rev. B 87, 075305 (2013).
22. B. O. Jahn, H. Ottesson, M. Galperin, and J. Fransson, ACS Nano 7, 1064 (2013).
23. K. Chen and Z. Zhang, Phys. Rev. Lett. 114, 126602 (2015).
24. S. Nakajima, M. Taguchi, T. Kubo, and Y. Tokura, Phys. Rev. B 92, 195420 (2015).
25. G. Tatara, Phys. Rev. B 94, 224412 (2016).
26. M. Moskalets, Scattering matrix approach to non-stationary quantum transport, (Imperial College Press, London, 2011).
27. K. Hashimoto, G. Tatara, and C. Uchiyama, Phys. Rev. B 96, 064439 (2017).
28. H.-P. Breuer and F. Petruccione, The Theory of Open Quantum Systems (Oxford University Press, Oxford, UK, 2002) Sec. 3.3.1.
29. This understanding is consistent with the fact the spin pumping effect is induced by the non-adiabatic component of effective gauge field 22.
30. G. Tatara, Physica E Low Dimens. Syst. Nanostruct. 106, 208 (2019).
31. A typical single molecule magnet studied in the molecular spintronics is octanuclear iron(III) oxo-hydroxo, [Fe₈O₂(OH)₁₂(tacn)]₆⁺ (in short, Fe₈). Following Ref. 22, the coupling strength between its lowest unoccupied molecular orbital (LUMO) and an external electron lead is $\Gamma \approx 0.0015$ meV. Therefore, the rate of relaxation by tunneling is estimated to be $\gamma \approx 0.36$ GHz. Assuming the precession of its magnetic core is excited by microwaves, the typical frequency of which is several tens of gigahertz, the precession frequency $\nu$ is more than ten times the relaxation rate $\gamma$.
32. M. Misiorny and J. Barnaś, Phys. Rev. B 76, 054446 (2007).
33. M. M. Wolf, J. Eisert, T. S. Cubitt, and J. I. Cirac, Phys. Rev. Lett. 101, 150402 (2008).
34. H.-P. Breuer, E.-M. Laine, and J. Piilo, Phys. Rev. Lett. 103, 210401 (2009).
35. H.-P. Breuer, J. Phys. B 45, 154001 (2012).
36. A. Rivas, S. F. Huelga, and M. B. Plenio, Phys. Rev. Lett. 105, 050403 (2010).
For $\lambda^\sigma = 0$, both the operator $\rho^{(A^\sigma)}(t)$ and the equation coincide with the reduced density matrix of the dot $\rho^{(0)}(t) \equiv \text{Tr}_l[W(t)]$, where $\text{Tr}_l$ is the trace taken over the lead and $W(t)$ is the density matrix of the total system, and to the master equation for $\rho^{(0)}(t)$ of the time convolutionless type.

We introduce a unit energy $\epsilon_u \equiv 2M$, a unit angular frequency $\omega_u \equiv 2M/\hbar$, and a unit time $t_u \equiv 2\pi/\omega_u$, and normalize energy, angular frequency, inverse temperature and time as $\bar{\omega} \equiv \omega/\omega_u$, $\bar{\omega} \equiv \omega/\omega_u$, $\bar{\epsilon} \equiv \epsilon/\epsilon_u$, $\bar{\beta} \equiv \beta/\beta_u$, and $\bar{t} \equiv t/t_u$, respectively.

The parameters satisfy conditions $\epsilon_d - M < \mu < \epsilon_d + M$ and $\beta^{-1} \lesssim 2M$, which are essential for spin pumping because, if they are not satisfied, either electrons do not transfer to the dot (as $\epsilon_d - M > \mu$) or spin-$\uparrow$ and spin-$\downarrow$ electrons of equal amounts flow onto the dot (for $\mu > \epsilon_d + M$).

The steady state $\rho^{st}$, satisfying $\xi^{(0)}(t \to \infty)\rho^{st} = 0$, is analytically obtained using the graphical method discussed in Ref.\[54\]. An analytical expression for the minimum model is provided in Appendix C of our previous paper Ref.\[27\].

As the maxima and minima are determined by the timing of the change of $\phi$ (or $\delta t$) and the Rabi period in the dot, $\hbar/2M$, they appear at the same frequencies in both the non-Markovian and Markovian results. For the Markovian analysis, we discussed in detail the oscillatory behavior in Ref.\[27\], Sec. 2.2. The discussion holds qualitatively for the non-Markovian analysis.