Supplementary Information

Magnetization dynamics induced by Rashba effect in ferromagnetic films

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1 Tight binding Hamiltonian

The tight binding Hamiltonian of the two-dimensional electron gas (2DEG) can be written as

$$\hat{H} = \sum_{\alpha=L,R} \hat{H}_\alpha + \hat{H}_C + \hat{H}_T, \tag{1}$$

where

$$\hat{H}_\alpha = \sum_{\mathbf{p}\sigma} \varepsilon_{\alpha\mathbf{p}} \hat{c}^\dagger_{\alpha\mathbf{p}\sigma} \hat{c}_{\alpha\mathbf{p}\sigma} + \sum_{\mathbf{pp}'\sigma} \hat{c}^\dagger_{\alpha\mathbf{p}\sigma} T_{\mathbf{pp}'} \hat{c}_{\alpha\mathbf{p}'\sigma}, \tag{2}$$

with

$$T_{\mathbf{pp}'} = -t_1 \mathbf{I}_s, \quad \text{for } \mathbf{p} = \mathbf{p}' + \hat{e}_x \text{ or } \mathbf{p} = \mathbf{p}' + \hat{e}_y. \tag{3}$$

Here, $\mathbf{I}_s$ is the identity matrix in the spin space. The Hamiltonian of the central region in the presence of the Rashba interaction is ($\hbar = 1$),

$$\begin{aligned}
\hat{H}_C &= \sum_{\mathbf{m}\sigma} \varepsilon_{\mathbf{m}} \hat{d}^\dagger_{\mathbf{m}\sigma} \hat{d}_{\mathbf{m}\sigma} - \frac{J}{2} \sum_{\mathbf{m}\sigma\sigma'} \hat{d}^\dagger_{\mathbf{m}\sigma} (\boldsymbol{\sigma} \cdot \hat{\mathbf{S}})_{\sigma\sigma'} \hat{d}_{\mathbf{m}\sigma'} + \sum_{\mathbf{mm}'\sigma\sigma'} \hat{d}^\dagger_{\mathbf{m}\sigma} T^{\sigma\sigma'}_{\mathbf{mm}'} \hat{d}_{\mathbf{m}'\sigma'} \\
&+ \frac{\gamma_e}{2} \sum_{\mathbf{m}\sigma\sigma'} \hat{d}^\dagger_{\mathbf{m}\sigma} (\boldsymbol{\sigma} \cdot \mathbf{B})_{\sigma\sigma'} \hat{d}_{\mathbf{m}\sigma'} + \gamma \sum_{\mathbf{m}\sigma} (\mathbf{S} \cdot \mathbf{B}) \hat{d}^\dagger_{\mathbf{m}\sigma} \hat{d}_{\mathbf{m}\sigma} + U^{\text{ani}}(\mathbf{S}) \sum_{\mathbf{m}\sigma} \hat{d}^\dagger_{\mathbf{m}\sigma} \hat{d}_{\mathbf{m}\sigma},
\end{aligned} \tag{4}$$

where $\hat{d}^\dagger_{\mathbf{m}}$ ($\hat{d}_{\mathbf{m}}$) creates (annihilates) an electron on site $\mathbf{m}$. The on-site potential $\varepsilon_{\mathbf{m}}$ describes the static local potential. In Eq. (4), $\hat{\mathbf{s}}_{\mathbf{m}} = \frac{1}{2} \psi^\dagger_{\mathbf{m}} \boldsymbol{\sigma} \psi_{\mathbf{m}}$ is the spin of itinerant electron in central region with $\psi_m = \begin{pmatrix} d_{\mathbf{m}\uparrow} \\ d_{\mathbf{m}\downarrow} \end{pmatrix}$, J is the exchange interaction describing the coupling of the electron levels with the single molecule magnet, γ and γ_e are the gyromagnetic ratio of the localized spin and itinerant electron spin, respectively, $\boldsymbol{\sigma}$ is the pauli matrix, $\hat{\mathbf{S}}$ is the localized spin which satisfies $[\hat{S}_x, \hat{S}_y] = i\hbar \hat{S}_z$, and $\mathbf{B}$ is the external magnetic field. The magnetization $\mathbf{M}$ is related to the localized spin via $\mathbf{S} = \mathbf{M} \mathcal{V} / \gamma$, with $\mathcal{V}$ the volume of the magnetization layer. The uniaxial anisotropy is parameterized by $U^{\text{ani}}(\mathbf{S}) = -DS_z^2$ [1]. The

nearest neighbor hopping $T_{mm'}^{\sigma\sigma'}$ accounts for the Rashba coupling which satisfies,

$$T_{mm'}^{\sigma\sigma'} = \begin{cases} -t_0\mathbf{I}_s - it_{SO}\sigma_y & \text{for } \mathbf{m} = \mathbf{m}' + \hat{e}_x, \\ -t_0\mathbf{I}_s + it_{SO}\sigma_x & \text{for } \mathbf{m} = \mathbf{m}' + \hat{e}_y. \end{cases} \tag{5}$$

Finally, the coupling between the quantum dot and the leads is described by,

$$\hat{H}_T = \sum_{\alpha\mathbf{pm}\sigma} [\hat{c}_{\alpha\mathbf{p}\sigma}^\dagger \tau_{\alpha\mathbf{pm}} \hat{d}_{\mathbf{m}\sigma} + h.c.], \tag{6}$$

where the coupling constant $\tau_{\alpha\mathbf{pm}}$ is

$$\tau_{L\mathbf{pm}} = -\tau_L\mathbf{I}_s, \quad \text{for } \mathbf{m} = \mathbf{p} + \hat{e}_x, \tag{7}$$

$$\tau_{R\mathbf{pm}} = -\tau_R\mathbf{I}_s, \quad \text{for } \mathbf{m} = \mathbf{p} - \hat{e}_x. \tag{8}$$

2 Magnetization dynamics in 2DEG system

From the Heisenberg equation of motion $\dot{\hat{\mathbf{S}}} = -i[\hat{\mathbf{S}}, \hat{H}]$, for the magnetization in the central region, we have

$$\frac{d\hat{\mathbf{S}}}{dt} = -J\hat{\mathbf{S}} \times \hat{\mathbf{s}}_C - \gamma\hat{\mathbf{S}} \times \mathbf{B} - \hat{\mathbf{S}} \times \partial_{\hat{\mathbf{S}}} U^{\text{ani}}(\hat{\mathbf{S}}). \tag{9}$$

Considering the saturation magnitude $\mathbf{S} = M\mathcal{V}\mathbf{m}/\gamma$ with the magnetization unit vector $\mathbf{m}$, Eq. (9) can be expressed as,

$$\frac{d\hat{\mathbf{m}}}{dt} = -J\hat{\mathbf{m}} \times \hat{\mathbf{s}}_C - \gamma\hat{\mathbf{m}} \times \mathbf{B} - \frac{\gamma}{M\mathcal{V}}\hat{\mathbf{m}} \times \partial_{\hat{\mathbf{m}}} U^{\text{ani}}(\hat{\mathbf{m}}), \tag{10}$$

where $\hat{\mathbf{s}}_C = \sum_{\mathbf{m}} \hat{\mathbf{s}}_{\mathbf{m}}$ is the electron spin in the central region. Now we neglect the fluctuations of the electron spin and magnetization for simplicity, namely, $\hat{\mathbf{s}}_C = \langle\hat{\mathbf{s}}_C\rangle$ and $\hat{\mathbf{m}} = \langle\hat{\mathbf{m}}\rangle$. Then the equation of motion can be turned into a Langevin equation for the expectation value $\mathbf{m}(t) = \langle\hat{\mathbf{m}}(t)\rangle$ of the magnetization[2],

$$\frac{d\mathbf{m}}{dt} = -\mathbf{m} \times [J\mathbf{s}_C + \gamma\mathbf{B} + \frac{\gamma}{M\mathcal{V}}\partial_{\mathbf{m}} U^{\text{ani}}(\mathbf{m})]. \tag{11}$$

From Eq. (11), we need to calculate the time dependence of the itinerant electron spin to study the magnetization dynamics. The electron spin $\mathbf{s}_C$ is defined as,

$$\mathbf{s}_C = -\frac{i}{2}\mathrm{Tr}[G^<(t,t)\boldsymbol{\sigma}], \tag{12}$$

where the detailed expression of lesser Green's function $G^<(t,t)$ is given in Eq. (29) shown below in Sec. 3. Using this expression, the electron spin can then be written as, $\mathbf{s}_C = \mathbf{s}_C^{(0)} + \mathbf{s}_C^{(1)}$, with the adiabatic term

$$\mathbf{s}_C^{(0)} = -\frac{i}{2}\int\frac{dE}{2\pi}\mathrm{Tr}[\boldsymbol{\sigma}G_f^<], \tag{13}$$

and the first order term

$$\mathbf{s}_C^{(1)} = -\frac{J}{4}\left(\frac{M\mathscr{V}}{\gamma}\right)\int\frac{dE}{2\pi}\mathrm{Tr}[G_f^<\boldsymbol{\sigma}G_f^rG_f^r\boldsymbol{\sigma} - G_f^aG_f^a\boldsymbol{\sigma}G_f^<\boldsymbol{\sigma}]\cdot\dot{\mathbf{m}}. \tag{14}$$

The retarded frozen Green's function G_f^r can be given in the adiabatic limit[3,4],

$$G_f^r(t,E) = \left[E - H_C(t) - \Sigma^r(E)\right]^{-1}, \tag{15}$$

where $H_C(t)$ is the Hamiltonian of the central region which depends on the instantaneous direction of the magnetization $\mathbf{m}(t)$. The advanced and lesser frozen Green's function defined in Sec. 3 is related to the retarded Green's function by $G_f^a = G_f^{r\dagger}$ and $G_f^< = G_f^r\Sigma^<G_f^a$.

In order to describe the magnetization in the spherical coordinate, the polar angle θ and the azimuthal angle ϕ are used to characterize the magnetization unit vector $\mathbf{m}(t)$. By using the relationship among unit vectors in Cartesian and spherical coordinates

$$\begin{aligned}
\hat{\mathbf{x}} &= \sin\theta\cos\phi\,\hat{\mathbf{r}} + \cos\theta\cos\phi\,\hat{\boldsymbol{\theta}} - \sin\phi\,\hat{\boldsymbol{\phi}}, \\
\hat{\mathbf{y}} &= \sin\theta\sin\phi\,\hat{\mathbf{r}} + \cos\theta\sin\phi\,\hat{\boldsymbol{\theta}} + \cos\phi\,\hat{\boldsymbol{\phi}}, \\
\hat{\mathbf{z}} &= \cos\theta\,\hat{\mathbf{r}} - \sin\theta\,\hat{\boldsymbol{\theta}},
\end{aligned} \tag{16}$$

and $d\hat{\mathbf{r}}/dt = \dot{\theta}\hat{\boldsymbol{\theta}} + \dot{\phi}\sin\theta\hat{\boldsymbol{\phi}}$, we can find the equations of dynamics from Eq. (11),

$$\frac{d\theta}{dt} = -\left[(Js_{C,x} + \gamma B_x)\sin\phi - (Js_{C,y} + \gamma B_y)\cos\phi\right], \tag{17}$$

$$\frac{d\phi}{dt}\sin\theta = -\left[(Js_{C,x} + \gamma B_x)\cos\theta\cos\phi + (Js_{C,y} + \gamma B_y)\cos\theta\sin\phi\right.$$

$$\left. - (Js_{C,z} + \gamma B_z - 2DS\cos\theta)\sin\theta\right]. \tag{18}$$

To summarize, with an initial magnetization orientation of $\mathbf{m}$, the Hamiltonian of the central region can be constructed by Eq. (4) and the corresponding frozen Green's functions can be obtained from Eq. (15). After calculating the itinerant electron spin using Eqs. (12)-(14), $\mathbf{m}$ can be updated through θ and ϕ in the spherical coordinate by the equations of dynamics [Eqs. (17)-(18)] to study the time evolution of magnetization.

3   Time-dependent Green's functions in instantaneous representation

In order to determine the electron spin $\mathbf{s}_C$ which is expressed in terms of $G^<(t,t)$, we now discuss the time dependent lesser Green's function in the instantaneous representation[4]. Using the equation of motion and the theorem of analytic continuation, one can obtain the differential form of Dyson equation on the real time axis[5],

$$\left[i\frac{\partial}{\partial t_1} - H(t_1)\right]G^r(t_1,t_2) - \int dt_3 \Sigma^r(t_1,t_3)G^r(t_3,t_2) = \delta(t_1 - t_2). \tag{19}$$

Here, G^r and Σ^r are the retarded Green's function and self energy, respectively. After taking the Fourier transform with respect to the time difference $\tau = t_1 - t_2$, we obtain,

$$\left[i\frac{\partial}{\partial t} + E - H(t)\right]G^r(t,E) - \int dt' \int \frac{dE'}{2\pi} e^{i(E-E')(t-t')}\Sigma^r(t,E')G^r(t',E) = I. \tag{20}$$

By using the wide band limit, namely, $\Sigma^{r,a}(t,E) = \Sigma^{r,a}$, the second term of Eq. (20) can be simplified

as,

$$\int dt' \int \frac{dE'}{2\pi} e^{i(E-E')(t-t')} \Sigma^r G^r(t',E)$$

$$= \int dt' e^{iE(t-t')} \int \frac{dE'}{2\pi} e^{-iE'(t-t')} \Sigma^r G^r(t',E) = \int dt' e^{iE(t-t')} \delta(t-t') \Sigma^r G^r(t',E)$$

$$= \Sigma^r G^r(t,E). \tag{21}$$

Hence Eq. (20) becomes

$$\left[i\frac{\partial}{\partial t} + E - H(t) - \Sigma^r \right] G^r(t,E) = I. \tag{22}$$

By introducing the definition of the retarded frozen Green's function,

$$[E - H(t) - \Sigma^r] G_f^r(t,E) = I, \tag{23}$$

and plugging it into Eq. (22), we find

$$G^r(t,E) = G_f^r(t,E) - iG_f^r(t,E)\dot{G}^r(t,E). \tag{24}$$

The corresponding advanced Green's function can be written as,

$$G^a(E,t) = G_f^a(E,t) + i\dot{G}^a(E,t)G_f^a(E,t). \tag{25}$$

Physically, the frozen Green function describes that the electrons adjust its experiencing potential instantaneously to the magnetization of the single-molecule magnet.

From the Keldysh equation, the lesser Green's function is given by,

$$G^<(t,t) = \int \frac{dE}{2\pi} G^r(t,E) \Sigma^<(E) G^a(E,t). \tag{26}$$

In order to calculate the lesser Green's function, we wish to make the adiabatic expansion for the Green's functions. Up to the first order in frequency, we can write the retarded (advanced) Green's

function from Eqs. (24) and (25) by

$$G^r(t,E) \approx G_f^r(t,E) - iG_f^r(t,E)\dot{G}_f^r(t,E), \tag{27}$$

$$G^a(E,t) \approx G_f^a(E,t) + i\dot{G}_f^a(E,t)G_f^a(E,t). \tag{28}$$

Substituting Eqs. (27) and (28) into Eq. (26), and keeping only the linear order, we have

$$G^<(t,t) \approx \int \frac{dE}{2\pi}[G_f^< - iG_f^r\dot{G}_f^r\Sigma^<G_f^a + iG_f^r\Sigma^<\dot{G}_f^aG_f^a], \tag{29}$$

where the frozen lesser Green's function is defined as,

$$G_f^< = G_f^r\Sigma^<(E)G_f^a, \tag{30}$$

with the abbreviation $G_f^r = G_f^r(t,E)$ and $G_f^a = G_f^a(E,t)$.

Notes and references

[1] N. Bode, L. Arrachea, G. S. Lozano, T. S. Nunner and F. von Oppen, *Phys. Rev. B*, 2012, **85**, 115440.

[2] J. L. García-Palacios and F. J. Lázaro, *Phys. Rev. B*, 1998, **58**, 14937–14958.

[3] N. Bode, S. V. Kusminskiy, R. Egger and F. von Oppen, *Beilstein J. Nanotechnol.*, 2012, **3**, 144.

[4] J. Splettstoesser, M. Governale, J. König and R. Fazio, *Phys. Rev. Lett.*, 2005, **95**, 246803.

[5] H. Haug and A.-P. Jauho, *Quantum kinetics in transport and optics of semiconductors*, Springer, Berlin, 1996.