Berry-Phase Oscillations of the Kondo Effect in Single-Molecule Magnets

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We show that it is possible to topologically induce or quench the Kondo resonance in the conductance of a single-molecule magnet ($S > 1/2$) strongly coupled to metallic leads. This can be achieved by applying a magnetic field perpendicular to the molecule easy axis and works for both full- and half-integer spin cases. The effect is caused by the Berry-phase interference between two quantum tunneling paths of the molecule's spin. We have calculated the renormalized Berry-phase oscillations of the Kondo peaks as a function of the transverse magnetic field as well as the conductance of the molecule by means of the poor man's scaling method. We propose to use a new variety of the single-molecule magnet Ni_4 for the experimental observation of this phenomenon.

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The quantum tunneling of the spin of single-molecule magnets (SMMs), such as Mn_{12} [1,2] and Fe_8 [3,4], has attracted a great deal of interest. These molecules have a large total spin, strong uniaxial anisotropy, and interact very weakly when forming a crystal. They have already been proposed for high-density magnetic storage as well as quantum computing applications [5]. Yet, there is much to explore in their fundamental properties. For instance, recent measurements of the magnetization in bulk Fe_8 samples (see Ref. [6]) have observed oscillations in the tunnel splitting $\Delta E_{m,m'}$ between states $S_z = m$ and m' as a function of a transverse magnetic field at temperatures between 0.05 K and $= 0.7$ K. Using a coherent spin-state path integral approach, it has been shown that this effect results from the interference between Berry phases carried by spin tunneling paths of opposite windings [7–9], a concept also applicable to transitions involving excited states of SMMs [10].

A new approach to the study of SMMs opened up recently with the first observations of quantized electronic transport through an *isolated* Mn_{12} molecule [11]. One expects a rich interplay between quantum tunneling, phase coherence, and electronic correlations in the transport properties of SMMs. It has been argued that the Kondo effect would only be observable for SMMs with half-integer spin [12] and therefore absent for SMMs such as Mn_{12}, Fe_8, and Ni_4, where the spin is integer. Here we show that this prediction is only valid in the absence of an external magnetic field. Remarkably, even a moderate transverse magnetic field topologically quenches the two lowest levels of a full-integer spin SMM, making them degenerate. The same Berry-phase interference also affects transport for SMMs with half-integer spin: In that case, sweeping the magnetic field will lead not to one but *a series* of Kondo resonances.

It is interesting to contrast the Kondo effect in a SMM with that observable in a lateral quantum dot with a single excess electron [13,14], in a single spin-1/2 atom [15], or in a single spin-1/2 molecule [16]. In those cases, at zero bias the Kondo effect is damped by an external a magnetic field because the degeneracy of the two spin states is lifted [17]. In the case of SMMs, the Berry-phase oscillations of the tunnel splitting $\Delta E_{m,m'}$ leads to oscillation of the Kondo effect as a function of $H_\perp$, the transverse magnetic field amplitude. This means that the Kondo effect is observable at zero bias for all values of $H_{\perp,0}$ such that $\Delta E_{m,m'}(H_{\perp,0}) = 0$. Notice that at a finite bias the Kondo effect in a quantum dot in the presence of a magnetic field of magnitude H can be restored by tuning the bias to $eV = \pm g\mu_B H$ (see Ref. [13]). For SMMs, however, the interference between the Berry phases accumulated by the molecule's spin makes the distance between the split Kondo peaks, which is equal to $eV = \pm\Delta E_{m,m'}$, oscillate as a function of $H_\perp$.

Consider a typical spin Hamiltonian of a SMM in an external transverse magnetic field $H_\perp$:

$$\mathcal{H}_{\rm spin} = -A_q S_{q,z}^2 + \frac{B_q}{2}(S_{q,+}^2 + S_{q,-}^2) + \frac{B_{4,q}}{3}(S_{q,+}^4 + S_{q,-}^4) + \frac{1}{2}(h_\perp^* S_{q,+} + h_\perp S_{q,-}), \quad (1)$$

where the easy axis is taken along z, $S_{q,\pm} = S_{q,x} \pm iS_{q,y}$, the integer index q denotes the charging state of the SMM, and $h_\perp = g\mu_B H_\perp$. Note that the transverse magnetic field lies in the xy plane. In this Hamiltonian, the dominant longitudinal anisotropy term creates a ladder structure in the molecule spectrum where the $|\pm m_q\rangle$ eigenstates of S_z are degenerate. The weak transverse anisotropy terms couple these states. The coupling parameters depend on the charging state of the molecule. For example, it is known that Mn_{12} changes its easy-axis anisotropy constant (and its total spin) from $A_0 = 56$ μeV ($S_0 = 10$) to $A_{-1} = 43$ μeV ($S_{-1} = 19/2$) and $A_{-2} = 32$ μeV ($S_{-2} = 10$) when singly and doubly charged, respectively [18].

The spin tunneling between the states $|m_q\rangle$ to $|-m_q\rangle$, with $|m_q| \le S_q$, can occur both clockwise and counter-

clockwise around the x axis. These two paths interfere with each other, which leads to Berry-phase oscillations [9,10]. Experiments with Ni_4 show that $B_{4,q=0} = -0.003$ K; i.e., $B_{4,0}$ is negative. In this case, in order to see the Berry-phase oscillation, the transverse magnetic field must be applied along angles that depend on the values of B_q and $B_{4,q}$ [10]. In Fig. 1 we show the Berry-phase oscillations of the tunnel splitting calculated for Ni_4 for two of such special orientations based on data from Ref. [19].

In order to show how these oscillations impact transport through the SMM, we first evaluate the Kondo effect for zero bias at the zero points, where the states $|m_q\rangle$ and $|-m_q\rangle$ are pairwise degenerate for all m_q. In order to consider the Kondo effect for SMMs, we need to add to the spin Hamiltonian in Eq. (1) the Kondo Hamiltonian [17]

$$\mathcal{H}_K = \sum_{k,s}\left[\xi_k + \frac{1}{2}(h_\perp^* s_+ + h_\perp s_-)\right]\psi_{ks}^\dagger\psi_{ks} + \sum_{m_q}\left[J_z^{(m_q)} s_z \Sigma_z^{(m_q)} + \frac{1}{2}J_\perp^{(m_q)}(s_+\Sigma_-^{(m_q)} + s_-\Sigma_+^{(m_q)})\right], \tag{2}$$

where $\mathbf{s} = \sum_{k,k',s,s'}\psi_{ks}^\dagger(\boldsymbol{\sigma}_{ss'}/2)\psi_{k's'}$ and the operators $\psi_{ks}^\dagger$ (ψ_{ks}) create (annihilate) electronic states in the leads with momentum k, spin s, and energy ξ_k. $\boldsymbol{\Sigma}^{(m_q)}$ is the pseudospin-1/2 operator acting on the states $|m_q\rangle$ and $|-m_q\rangle$. We define $\Sigma_\pm^{(m_q)} = \Sigma_x^{(m_q)} \pm i\Sigma_y^{(m_q)} = |\pm m_q\rangle\times\langle\mp m_q|$ and $\Sigma_z^{(m_q)} = (|m_q\rangle\langle m_q| - |-m_q\rangle\langle -m_q|)/2$. The spin-flip terms in Eq. (2) induce the Kondo resonance. The exchange part, $\mathcal{H}_{\rm ex}$ [the second term on the right-hand side of Eq. (2)] can be derived from a generalized Anderson's impurity model that takes into account the charging dependence of the total spin of the SMM. When the charging energy U is much larger than the tunneling matrix element t between the leads and the SMM, the exchange coupling can be derived in second-order perturbation theory by means of a Schrieffer-Wolff transformation [20], yielding

$$\begin{aligned} J_\perp^{(m_q)} &= \frac{2t^2}{U}\left[\frac{\Delta E_{m_{q-1},-m_{q-1}}}{\frac{U}{2}+\Delta E_{m_{q-1},-m_{q-1}}} + \frac{\Delta E_{m_{q+1},-m_{q+1}}}{\frac{U}{2}+\Delta E_{m_{q+1},-m_{q+1}}}\right] \\ &\approx 4t^2(\Delta E_{m_{q-1},-m_{q-1}} + \Delta E_{m_{q+1},-m_{q+1}})/U^2, \\ J_z^{(m_q)} &= \frac{4t^2}{U}\left[\frac{U+\Delta E_{m_{q-1},-m_{q-1}}}{\frac{U}{2}+\Delta E_{m_{q-1},-m_{q-1}}} + \frac{U+\Delta E_{m_{q+1},-m_{q+1}}}{\frac{U}{2}+\Delta E_{m_{q+1},-m_{q+1}}}\right] \\ &\approx 8t^2/U, \end{aligned} \tag{3}$$

where U is the charging energy. Notice that the coupling is antiferromagnetic and $J_z^{(m_q)} \gg J_\perp^{(m_q)}$. Since $J_\perp^{(m_q)}$ depends on the tunnel splittings $\Delta E_{m_{q-1},-m_{q-1}}$ and $\Delta E_{m_{q+1},-m_{q+1}}$ (which correspond to one charge added and removed from the SMM, respectively), the Kondo exchange coupling is strongly anisotropic, which confirms the result obtained in Ref. [12].

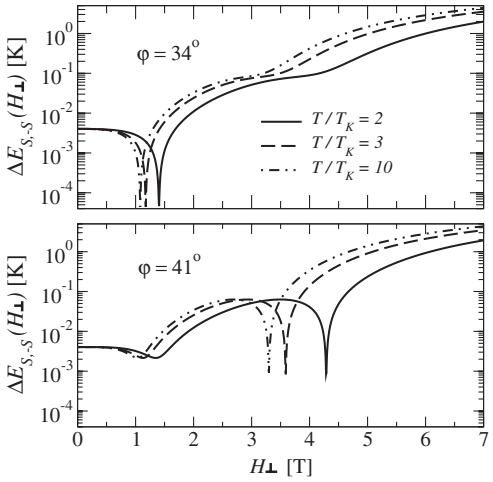

FIG. 1. Berry-phase oscillations of the spin tunnel splitting in the Ni_4 spin cluster of Ref. [19] at different temperatures ($S = 4$). The anisotropy constants are $A = -1.33$ K, $B = 0.034$ K, and $B_4^4 = -0.003$ K. $\Delta E_{S,-S}$ is calculated by an exact diagonalization for $\varphi = 33.5°$ and $\varphi = 40.75°$. As T approaches T_K, the renormalization of the g factor increases, an effect that can be verified experimentally. The bare g factor is $g = 2.2$ for Ni_4.

The transverse anisotropy terms in Eq. (1) mix the states $|m_q\rangle$. Since $|A| \gg |B|$, $|B_4|S^2$, the eigenstates are nondegenerate and of the form $|m_q^{(\pm)}\rangle = (|m_q\rangle \pm |-m_q\rangle)/\sqrt{2}$. However, at the zero points the SU(2) symmetry is restored, yielding degenerate eigenstates of the form $|\pm m_q\rangle$. The Kondo Hamiltonian in Eq. (2) opens up transition paths between pairs of degenerate states $|m_q\rangle$ and $|-m_q\rangle$ and these paths depend on temperature. This leads in general to multipath (but single-channel) Kondo correlations at $T > T_K$. At $T = 0$, however, only the $|\pm S\rangle$ states contribute to the Kondo effect. The unusual feature of Eq. (2) is that the total spin is not conserved. After a spin-flipping event, the excess angular momentum $L = 2m_{q-1}\hbar$ must be absorbed by orbital (and possibly nuclear) degrees of freedom in the SMM and then be transferred to the molecule as a whole. Since the kinetic energy of a rotation of tens of $\hbar$ corresponds to a few mK for a typical SMM, the excess orbital angular momentum will be relaxed by thermal fluctuations in the metallic contacts. The critical assumption we make is that the excess angular momentum is transferred from spin to orbital (and possibly nuclear) degrees of freedom fast enough to allow for the Kondo state to be formed.

Our analysis employs the standard poor man's scaling [21] to renormalize the effective exchange coupling constants J_z, J_+, J_-, and the g factor. In order to make the discussion self-contained, we present the main steps of the derivation. We start by calculating the renormalization flow at the zero points where the Kondo effect is observable for zero bias. It is reasonable to assume that

$h_{x,0,n} \gg T_K$, except when $n = 0$ for half-integer spins. The total Hamiltonian for the combined SMM and leads system reads (we will suppress the index q hereafter)

$$\mathcal{H}_{\rm tot} = \sum_{m\geq 0}\Big[\epsilon_m(\Sigma_z^{(m)})^2 + \frac{1}{2}\eta^{(m)}(\tilde{h}_\perp^*\Sigma_+^{(m)} + \tilde{h}_\perp\Sigma_-^{(m)})\Big] + \sum_{k,s}\xi_k\psi_{ks}^\dagger\psi_{ks} + \mathcal{H}_{\rm ex}, \quad (4)$$

where ϵ_m is the eigenvalue of $|m\rangle$ and $|-m\rangle$ at the zero points, $|\tilde{h}_\perp|$ is the effective Zeeman splitting between $(e^{-i\varphi/2}|m\rangle + e^{i\varphi/2}|-m\rangle)/\sqrt{2}$ and $(e^{-i\varphi/2}|m\rangle - e^{i\varphi/2}|-m\rangle)/\sqrt{2}$, and $\eta^{(m)} = 1 - \rho_0 j_\perp^{(m)}/2$ due to the Knight shift, with ρ_0 denoting the density of states of the itinerant electrons. The Zeeman term for the itinerant electrons is absent in Eq. (4) because at finite values of $h_{\perp,0,n}$ one has to cut the edges of the spin-up and spin-down bands in the leads to make them symmetric with respect to the Fermi energy [17]. Let us call D the resulting band width. The Hamiltonian remains invariant under renormalization group transformations. Using a one-loop expansion (second-order perturbation theory), we obtain the flow equations

$$\frac{dJ_\pm^{(m)}}{d\zeta} = -2\rho_0 J_\pm^{(m)}J_z^{(m)}, \qquad \frac{dJ_z^{(m)}}{d\zeta} = -2\rho_0 J_+^{(m)}J_-^{(m)}, \quad (5)$$

$$\frac{d\eta^{(m)}}{d\zeta} = \frac{\rho_0^2}{2}(J_+^{(m)} + J_-^{(m)})J_z^{(m)}, \quad (6)$$

where $\zeta = \ln(\tilde{D}/D)$ and $\tilde{D}$ is the rescaled band width. Dividing Eqs. (5) and integrating by parts gives $(J_z^{(m)})^2 - (J_\perp^{(m)})^2 = C^{(m)}$, where $C^{(m)}$ is a positive constant [21]. The exchange coupling constants always flow to an antiferromagnetic state because $J_z^{(m)} > 0$, but at the end of the flow the coupling tends to become isotropic. Solving Eqs. (5) yields

$$\frac{1}{2\rho_0\sqrt{C^{(m)}}}\operatorname{arctanh}\left(\frac{\sqrt{C^{(m)}}}{J_z^{(m)}}\right) = \ln\left(\frac{\tilde{D}}{T_K}\right). \quad (7)$$

The solution for $J_\perp^{(m)}$ is determined by $J_z^{(m)} = \sqrt{(J_\perp^{(m)})^2 + C^{(m)}}$. The flow stops at $\tilde{D} \approx \omega \sim T > T_K$. The result of the flow for η is presented in Fig. 2.

We are now ready to calculate the linear conductance through the SMM, which is given by the formula [17]

$$G = G_0\int d\omega\left(-\frac{df}{d\omega}\right)\frac{\pi^2\rho_0^2}{16}\frac{\sum_m e^{-\epsilon_m/T}|\mathcal{A}^{(m)}(\omega)|^2}{\sum_m e^{-\epsilon_m/T}}, \quad (8)$$

where G_0 is the classical (incoherent) conductance of the molecule. At the end of the flow the transition amplitude $\mathcal{A}^{(m)}$ can be calculated in first-order perturbation theory, and one finds that $\eta_{\tilde{D}\approx\omega}^{(m)} = 1 - \rho_0 J_{\perp,\omega}^{(m)}/2$ and

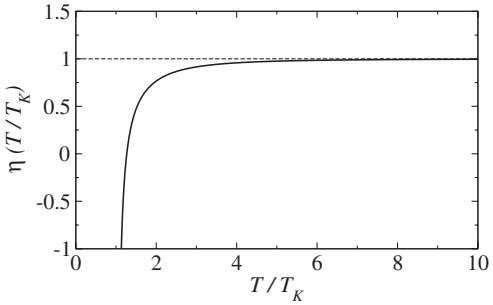

FIG. 2. The renormalization of the g factor due to the Knight shift. As an estimate, we used $\rho_0 J_\pm = \rho_0 J_z = 0.15$ [15].

$$\mathcal{A}_{\tilde{D}\approx\omega}^{(m)} = J_{\perp,\omega}^{(m)} = \sqrt{C^{(m)}}\left[\frac{(\omega/T_K)^{2\rho_0\sqrt{C^{(m)}}}}{(\omega/T_K)^{4\rho_0\sqrt{C^{(m)}}} - 1}\right]. \quad (9)$$

Substituting ω by T and inserting Eq. (9) into (8), one finds that the conductance diverges as $T \to T_K$, signaling the onset of the Kondo effect. Since $C^{(m)} > 0$ the singularity in Eq. (9) differs from the usual logarithmic behavior. The Kondo effect causes a fundamental change in the SMM behavior: All the zero points of the Berry-phase oscillation get rescaled by the g-factor renormalization: $b_{\perp,0,n}^{(m)} = h_{\perp,0,n}/\eta_{\tilde{D}\approx T}^{(m)}$. Thus, the zero points become dependent on the contributing states $|m\rangle$ and $|-m\rangle$. This result indicates that the period of the Berry-phase oscillations becomes temperature dependent as T is lowered toward T_K. Remarkably, the scaling equations can be checked experimentally by measuring the renormalized zero points of the Berry phase. Moreover, due to the scale invariance of the Kondo effect, the period of oscillations should follow a universal function of T/T_K.

The necessary conditions for observing these oscillations are a large enough tunnel splitting and a strong coupling between the SMM and the leads. In regard to the former, a new Ni_4 single-molecule magnet with $S = 4$ has been synthesized [19] with $\Delta E_{S,-S} \sim 0.01$ K or larger, depending on $h_\perp$. However, the two recent reports of transport through a SMM [11] show that the electrical contacts between the SMM and the leads is rather poor and would need to be improved in order to bring T_K to accessible values.

Let us now study the linear conductance for nonzero bias, $eV = \mu_L - \mu_R \neq 0$. We focus on the low-temperature regime, where we can substitute $-df/d\omega$ by $[\delta(\omega + eV/2) - \delta(\omega - eV/2)]/2$ in Eq. (8). We consider the situation where one moves from the zero points $b_{\perp,0,n}$ to $b_\perp = b_{\perp,0,n} + \Delta b_\perp$, with $\Delta b_\perp = \eta_{b_{\perp,0,n}}\Delta h_\perp$. If $|eV| \ll \Delta E_{m,-m}(b_\perp) \ll T_K$, the transmission amplitude is well approximated by Eq. (9). On the other hand, for $|eV| \gg T_K \gg \Delta E_{m,-m}(b_\perp)$ the transmission amplitude is given by $\mathcal{A}_{eV} = J_{\perp,eV}$. For the case $|eV| \sim \Delta E_{m,-m}(b_\perp) \gg T_K$ we can expand $\mathcal{A}_{\tilde{D}\approx\max\{T,\Delta E_{m,-m}(b_\perp)\}}$ up to second order in perturbation theory at the flow end,

yielding

$$\mathcal{A}_{\tilde{D}}^{(m)}(\omega) = J_{\perp,\tilde{D}}^{(m)} + \rho_0 J_{\perp,\tilde{D}}^{(m)} J_{z,\tilde{D}}^{(m)} \ln\left|\frac{\omega + \tilde{D} - eV/2}{\omega - \tilde{D} + eV/2}\right|, \tag{10}$$

where the integration limits account for the asymmetric cut of the bands. Keeping terms up to third order in $J_{\tilde{D}}^{(m)}$ and combining the results for zero and nonzero bias, we obtain

$$\frac{G}{G_0} = \frac{\pi^2 \rho_0^2}{16} J_{\perp,\tilde{D}}^2 \left[\delta_{eV,0} + \rho_0 J_{z,\tilde{D}} \ln\left(\frac{\Delta E_{m,-m}}{||eV| - \Delta E_{m,-m}|}\right)\right], \tag{11}$$

which agrees with Ref. [22]. The two split Kondo peaks appear at $|eV| = \Delta E_{m,-m}(b_\perp)$. Thus, the distance between the two peaks oscillates with magnetic field, following the renormalized periodic oscillations of the tunnel splitting $\Delta E_{m,-m}(b_\perp)$.

These results can be extended to the strong-coupling Kondo regime, namely, at $T = 0$, where only the two lowest-lying states $|S\rangle$ and $|-S\rangle$ contribute to the Kondo effect. Similarly to the spin $S = 1/2$ case, our calculations yield $G(T = 0) = \frac{G_0}{2}\sum_s \sin^2\delta_s = G_0$ at the zero points of the Berry-phase oscillation, where $|\delta_s| = \pi/2$ is the scattering phase shift in the unitary limit. In order to find the zero points of the Berry-phase oscillation, one must employ a more accurate approach, such as the numerical renormalization group technique [23]. However, since the ground state of the Kondo model given by Eq. (2) has $S_{\text{Kondo}} = S_q - 1/2$ due to the spin screening provided by the itinerant electrons, we conclude that the spin parity of the SMM effectively changes from even to odd or from odd to even when one goes from the high- to the low-temperature Kondo regimes. This means that, e.g., Ni_4 should behave as if $S_{\text{Kondo}} = 7/2$ at $T \ll T_K$.

To help guide the experimental effort on this problem, we provide some estimates for the Kondo temperature using the expression $T_K = D\exp[-\text{arctanh}(\sqrt{C^{(S)}}/J_z)/2\rho_0\sqrt{C^{(S)}}]$ derived from Eq. (7). Using $\rho_0 J_z = 0.15$ (similar to Ref. [16]) and setting Δ equal to the level spacing in the SMM, $D = |A[S^2 - (S-1)^2]| = 9.3$ K, we obtain $T_K = 1.2$ K in Ni_4 for the tunneling between the ground states $m = S = 4$ and $m = -S = -4$. The two crucial ingredients for the experimental observation in SMMs are (i) a large spin tunnel splitting and (ii) a large tunneling amplitude between the leads and the SMM. The first requirement is satisfied by Ni_4. The second one remains an experimental challenge. In the case of Mn_{12}, $\Delta E_{S,-S}(H_\perp \sim 0) \sim 10^{-10}$ K for the ground state tunneling, which leads to a negligible small Kondo temperature T_K. However, $\Delta E_{4,-4}(H_\perp \sim 0) \sim 0.01$ K, which leads to $T_K \sim 1$ K. Unfortunately, since the excited levels $m = \pm 4$ are only populated at temperatures of about 1 K, the levels $m = \pm 4$ cannot be resolved by the electrons in the leads.

In summary, we have shown that the Kondo effect in single-molecule magnets attached to metallic electrodes is a nonmonotonic (possibly periodic) function of a transverse magnetic field. This behavior is due to Berry-phase oscillations of the molecule's large spin. The period of these oscillations is strongly renormalized near the Kondo temperature and should follow a universal function of temperature that can be accessed experimentally. We argue that a newly synthesized family of Ni_4 SMMs meets the requirements for such experiment.

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