Low-temperature relaxation of an anisotropic semiclassical spin: tunnel flipping

A. S. Ioselevich
L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR

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The probability for the flipping of a spin $s \gg 1$ (with an easy-axis anisotropy) due to a transverse field or an interaction with phonons is calculated. The flipping is interpreted as a tunneling through a magnetic-anisotropy barrier. The behavior as a function of the temperature and the longitudinal field is derived.

We assume that the spin Hamiltonian of a magnetic impurity in a nonmetallic matrix is axisymmetric and corresponds to the easy-axis case: $V_0(\hat{\mathbf{s}}) = Wf(s_z/s)$, where W is the height of the "barrier," and $f(-1) = 0$ (Fig. 1). At a low temperature a spin can actually be in only two states: $\psi_-(s_z = -s)$ and $\psi_+(s_z = s)$. Transi-

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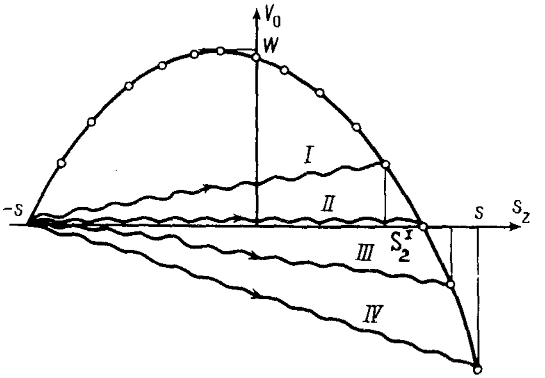

FIG. 1. Diagram of the tunneling transitions. In the case of a transverse field, only elastic transition II is possible; in the case of an interaction with phonons, the most favorable process is inelastic transition IV, with the maximum release of energy.

tions between these states are possible if there is a perturbation V_{int}, e.g., a field $\mathbf{H}\perp z$ or an interaction with phonons of the reservoir. If V_{int} is linear in $s_{\pm}$, a transition from ψ_- to ψ_+ requires that the spin go through all of the intermediate virtual states in succession: $-s\to -s+1\to\cdots\to s$. For a quadratic V_{int}, every other intermediate state is entered: $-s\to -s+2\to\cdots$. This "motion" along the coordinate s_z is reminiscent of a tunneling through a potential barrier. In ordinary tunneling, different spatial points are coupled by a kinetic-energy operator which contains gradients; in our case, the possibility of a motion comes from the perturbation: As the perturbation becomes weaker, the spin becomes "heavier" and thus less successful in tunneling. It is legitimate to speak in terms of a tunneling if the number of intermediate states is large: $s\gg 1$. If V_{int} is small, the transition amplitude A_{+-} can in principle be derived in a high-order ($\sim s$) perturbation theory. A picture of the tunneling comes from the $s\gg 1$ asymptotic method for carrying out this calculation. Here we will consider three types of V_{int}: a weak transverse field H_x, an interaction with an oscillator, and an interaction with acoustic phonons.

1. Transverse field. The semiclassical spin is described by the Lagrangian

$$L = L_s^{(0)} - V_{int}, \qquad L_s^{(0)} = -s\cos\theta\,\dot{\varphi} - Wf(\cos\theta) \tag{1}$$

(e.g., Ref. 1), where $\hbar=1$, $s_z = s\cos\theta$, and $s_{\pm} = s\sin\theta e^{\pm i\varphi}$. In the case of a transverse field we would have $V_{\text{int}} = \Delta_x s\sin\theta\cos\varphi$, where $\Delta_x = g_\perp\mu_0 H_x$, $g_\perp$ is the transverse g-factor, and μ_0 is the Bohr magneton. The simplified action is

$$S_0 = \int\frac{\partial L}{\partial\dot{\varphi}}\,d\varphi = -s\int\cos\theta\,d\varphi = \int\varphi\,d(s\cos\theta), \tag{2}$$

from which we see that we have $\varphi = p_{s_z}$; i.e., the precession angle is the canonical momentum which is the conjugate of S_z. To find the tunneling action, which determines the transparency, we switch to the imaginary time $t = i\tau$ and the imaginary angle $\varphi = \pi + i\psi$. Eliminating ψ with the help of the conservation law for the energy $E = V_0 + V_{\text{int}}$, we find

$$\tilde{S}_0(E) = -iS_0(E) = \int_{s_1(E)}^{s_2(E)} \tilde{p}_{s_z}(E, s_z)ds_z \tag{3}$$

$$\tilde{p}_{s_z}(Es_z) = \psi = \operatorname{arccosh}\left(\frac{V_0(s_z) - E}{\Delta_x\sqrt{s^2 - s_z^2}}\right) \tag{4}$$

where $s_{1,2}$ are turning points. The semiclassical treatment is valid under the condition $d\tilde{p}_{s_z}/ds_z \ll \tilde{p}_{s_z}^2$; using (4), we then find the condition $\Delta_x \ll W/s$, which agrees with the condition for the applicability of the perturbation theory. Setting $H_z = 0$ [i.e., $f(x) = f(-x)$], we can easily evaluate the integral in (3). For $E = 0$ we find

$$A_{+-} \propto \exp(-\tilde{S}_0) = (cs\Delta_x / W)^{2s}$$
$$c = (e/4)\exp\left\{\int_0^1 \ln\left(\frac{1-\kappa^2}{f(\kappa)}\right)d\kappa\right\}. \tag{5}$$

The result $A_{+-} \propto (s\Delta_x/W)^{2s}$ is obvious from the perturbation-theory standpoint; the only nontrivial matter is determining the number c. We can also calculate the coefficient of the exponential function and take the field H_z into account, but we will not reproduce these calculations here.

The transparency is thus given by the customary expression, (3). The only unusual aspect is the logarithmic, rather than square-root, dependence of the momentum on the potential energy: $\tilde{p}_{s_z} \approx \ln(W/s\Delta_x)$. This is a general situation for problems in which the action changes in discrete jumps $\delta S > 1$ (Ref. 2).

2. Interaction with an oscillator. We consider an oscillator **Q** with a mass m_0 and a frequency ω, which has a temperature T, and which is interacting with a spin through $V_{\text{int}} = U_0(Q_\alpha s_\alpha)^2$ (α runs over the values x,y; the interaction with Q_z is inconsequential). We take the longitudinal field H_z into account and thus write $V_0(s) = -2s\Delta_z < 0$, $\Delta_z = g_\parallel\mu_0 H_z$ (Fig. 1). Introducing the variables $Q_\pm = Qe^{\mp i\alpha'}$, and switching to the imaginary time $t = i\tau$ and the imaginary angles $\varphi = i\psi$, and $\alpha' = i\alpha$, we find the Lagrangian of the system

$$L = L_s^{(0)} + \frac{m_0}{2}[Q^2(\dot{\alpha}^2 - \omega^2) - \dot{Q}^2] - U_0 s^2 \sin^2\theta \cosh^2(\psi + \alpha). \tag{6}$$

To calculate ω_{+-}, the transition probability averaged over the initial states, we need to find the total action $\tilde{S}$ corresponding to a tunneling time of $1/2T$, with $w_{+-} \propto e^{-2\tilde{S}}$. We assume

$$\Delta_z, \omega, T \ll W/s; \quad 2\tilde{S} < W/T. \tag{7}$$

We conclude from (7) that (1) the field perturbs the barrier relatively weakly, (2) only the oscillator, not the spin, can be excited in the initial state, and (3) the tunneling process is favored over a purely activation process: $w_{+-} > e^{-(W/T)}$.

Putting Lagrangian (6) in dimensionless form, we find the characteristic values

$\tau_0 \sim s/W$, $Q_0 \sim s(m_0 W)^{-1/2}$ and $\psi + \alpha \sim \ln(W m_0^{1/2}/U_0^{1/2} s^2)$. The temperature-field dependence of w_{+-} is determined, by virtue of (7), by the oscillatory part of the action and can easily be separated out. As a result, we find

$$w_{+-} \propto \left(\frac{U_0 s^5 \omega}{W^3 m_0}\right)^{2s} \begin{cases} \exp(-\epsilon_A/T) & (2T^* < \omega) \\ \left(\frac{2T^*}{\omega}\right)^{2s} & (2T^* > \omega) \end{cases}, \quad (8)$$

where $T^* \approx \max(T, \Delta_z/2)$ and $\epsilon_A = s(\omega - \Delta_z)$. Structually, expression (8) is a result of a perturbation theory. An important point is that under the condition $\Delta_z < \omega$ the transition is not a pure tunneling, and in the limit $T \to 0$ it corresponds to a finite activation energy ϵ_A. Why?

Lagrangian (6) leads to the conservation of the projection of the total angular momentum of the system, $M = s_z + m_0 Q^2 \dot{\alpha}$; consequently, upon a flipping of the spin, angular momentum is transferred to the oscillator. If the spin tunnels through the barrier at point s_2 (Fig. 1), the oscillator acquires an angular momentum $\Delta M = -s - s_2$ and an energy $\Delta E = -V_0(s_2)$. If $\Delta E > 0$, $N_0 = \Delta E/\omega$ phonons can be emitted; they will carry off an angular momentum $(-1)N_0$. If $N_0 = < |\Delta M|$, the remaining angular momentum $\Delta' M = \Delta M + N_0$ can be transferred only to existing thermal phonons, each of which, through a flipping of its "spin" (from 1 to -1), carries off an angular momentum of -2. The smallest necessary number of thermal phonons is therefore $N_T = |\Delta' M|/2$, and their total energy is $N_T \omega = 1/2[(s+s_2)\omega + V_0(s_2)]$. How do we find the optimum value of s_2? The activation probability $\exp(-N_T \omega/T)$ increases with s_2, while the amplitude A_{+-} decreases {since the necessary order of the perturbation theory, $[(s+s_2)/2]$, increases}. It is easy to show that under condition (7) the first of these tendencies is dominant, so that we have $s_2 = s$, the process takes path IV (Fig. 1), and we have $N_T \omega = \epsilon_A$. If $\Delta_z > \omega$, then we have $N_T = 0$, since the amount of energy released is sufficient for a removal of angular momentum only through emission, and the process becomes a pure tunneling. The optimum value of s_2 remains equal to s up to $\Delta_z \sim W/2\widetilde{S} \gg \omega$, and the following qualitative picture holds: In the initial state (χ_-) the oscillator is not excited, while in the final state (χ_+) its energy is ΔE, and its angular momentum is ΔM. The spin flips at $Q \sim Q_0$, so that we have $w_{+-} \propto |\chi_-^*(Q_0)\chi_+(Q_0)|^2 \propto (Q_0\sqrt{m_0 \Delta E'})^{2|\Delta M|} \propto \Delta_z^{2S}$. Similar arguments lead to the behavior $w_{+-} \propto (2T)^{2S}$ at $2T \gg \Delta_z, \omega$.

3. Acoustic phonons. We consider isotropic long-wave acoustic phonons which are interacting with a spin through $V_{\text{int}} = U u_{\alpha\beta}(0) S_\alpha S_\beta$, where $u_{\alpha\beta}(0)$ is the strain energy at an impurity site, and U is the spin-strain interaction. The potential V_{int} is governed primarily by the longitudinal and "electric transverse" quadrupole phonons with $j = 2$, $m = \pm 2$ (j is the total angular momentum of a phonon; m is its projection; and the terminology is the standard terminology in work with spherical phonons; see Ref. 3). These contributions are comparable in magnitude. For simplicity, we will retain only the longitudinal mode. We introduce $Q^{(l)}_{2,\pm 2,k} = \pm Q_k \exp(\pm 2i\alpha'_k)$, where $Q^{(l)}_{jmk}$ is the amplitude of the jmk longitudinal phonon. The Lagrangian of the system (in terms of the imaginary time and angles) is

$$L = L_s^{(0)} + \int \frac{k^2 dk}{2\pi^2} \left\{ \frac{\rho}{2} (Q_k^2 (4\dot{\alpha}_k^2 - v^2 k^2) - \dot{Q}_k^2) - \sqrt{\frac{2}{15}} Uks^2 \sin^2\theta \cosh(2(\psi + \alpha_k)) \right\}, \quad (9)$$

where ρ is the density, and v is the sound velocity. Putting (9) in dimensionless form, we find $Q_0 \sim s(W\rho k_0^3)^{-1/2}$, and $\psi + \alpha \sim 1/2 \ln[(\rho W^3/k_0^5)^{1/2}/Us^2]$, $vk_0 \sim T^*$. As a result, we find

$$w_{+-} \propto (Us^{5/2} T^{*3}/W^2 \rho^{1/2} v^{5/2})^{2s} \quad (10)$$

The dependence on T and Δ_z is always a power-law dependence here, since there are phonons at arbitrarily low frequencies, and we can lower the activation energy without bound, in the process giving up something in terms of the strength of the interaction and the phase space. The optimum situation is reached at $\omega \sim T^*$, and a functional dependence $(T^*)^{6s}$ is generated.

In summary, in a weak longitudinal field $H \ll H^* = 2T/g_\parallel \mu_0$ the time required for the magnetization of a sample with impurities, $\tau_\parallel(H)$, and the time required for its demagnetization, $\tau_\parallel(0)$, are equal to each other and are very strong functions of T. At $H \gtrsim H^*$, the time $\tau_\parallel(H)$ falls off sharply and is independent of T: $\tau_\parallel(H)/\tau_\parallel(0) \sim (H^*/H)^{6s}$. The mechanism proposed here can describe the relaxation of the angular momentum of not only a single impurity with $s \gg 1$ but also of single-domain magnetic clusters.

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