Quantum Dynamics of Domain Walls in Molecular Magnets

D. A. Garanin and E. M. Chudnovsky
Lehman College, City University of New York

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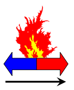
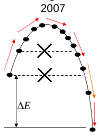
Magnetic Burning: From thermal to quantum



Deflagration 2007



- controlled by temperature

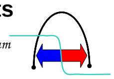
Model:

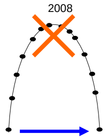
$$\hat{H} = -D\hat{S}_z^2 - g\mu_B H_z \hat{S}_z + \hat{H}'$$

Create barrier
Bias wells; Tune resonances
Cause tunneling

$$+\hat{H}_{\mathrm{DDI}} + \hat{H}_{\text{spin-phonon}}$$

Domain wall 2008





- controlled by external + dipolar field

Our main point:

Tunneling relaxation in MMs can occur via self-induced resonance tuning of the dipolar field leading to a propagating front of Landau-Zener transitions.

- For large external bias this is quantum deflagration (with heating added)
- For small external bias this is propagation of quantum domain walls

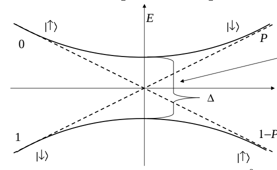
Here we concentrate on quantum domain walls near the zero-field tunneling resonance.

Domain walls and the underlying ordering*) are due to the DDI.

*) Investigated and observed on several MMs (Fernandez & Alonso; Martinez Hidalgo, Chudnovsky and Aharony; Luis et al; Morello et al; Belesi, Borsa and Powell)

Landau-Zener Effect

$$\hat{H}(t) = -\tfrac{1}{2}W(t)\hat{\sigma}_z - \tfrac{1}{2}\Delta\hat{\sigma}_x$$



$$|\psi\rangle_t = C_\downarrow(t)|\downarrow\rangle + C_\uparrow(t)|\uparrow\rangle, \qquad P(t) = |C_\downarrow(t)|^2$$

staying probability: $P(t=\infty) = e^{-\varepsilon}, \quad \varepsilon = \dfrac{\pi\Delta^2}{2\hbar v}$ (1932) — Landau, Zener, Stueckelberg, Majorana

$W(t) = vt$

experimental observation on Fe-8: Wernsdorfer *et al*, J. Appl. Phys.-2000

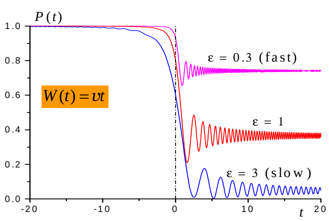
"Magnetization": $\boldsymbol{\sigma} = \langle\psi|\hat{\boldsymbol{\sigma}}|\psi\rangle \qquad \sigma_z(t) = |C_\uparrow|^2 - |C_\downarrow|^2 = 1 - 2P(t)$

Landau-Lifshitz equation for the LZ effect:
Is equivalent to the Schrödinger equation!

$$\dot{\boldsymbol{\sigma}} = \frac{1}{\hbar}[\boldsymbol{\sigma}\times\mathbf{A}(t)], \qquad \mathbf{A}(t) = \mathbf{e}_z W(t) + \mathbf{e}_x\Delta$$

Connection: LZ effect – DW dynamics

Time dependence:

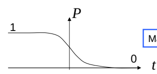


Time-nonlinear LZ problems

- **Inverse Landau-Zener problem**

given $P(t) \xrightarrow{?} W(t)$

Exact solution:
Garanin & Schilling, EPL-2002



Manipulation of quantum states

- **Self-consistent LZ problem – $W(t)$ created by transitions of other spins**

Example: Domain walls in ferromagnets. Self-consistent time-dependent field on a spin results in a complete LZ transition. Exact analytical Walker solution for the dissipationless DW motion – Döring mass of the DW

Moving DW ↔ LZ front

In general, LZ transition is incomplete and there are excitations behind the front

Formulation of the model

Equation of motion:

Use density-matrix equation for a pseudospin ½ coupled to bath. With

$$\boldsymbol{\sigma} = \mathrm{Tr}(\hat{\rho}\hat{\boldsymbol{\sigma}}), \qquad \hbar\boldsymbol{\omega}_0 = \mathbf{A}(t) = \mathbf{e}_z W(t) + \mathbf{e}_x\Delta$$

one obtains

$$\dot{\boldsymbol{\sigma}} = [\boldsymbol{\sigma}\times\boldsymbol{\omega}_0] - \frac{\Gamma}{2}\left(\boldsymbol{\sigma} - \frac{\boldsymbol{\omega}_0\bullet\boldsymbol{\sigma}}{\omega_0^2}\boldsymbol{\omega}_0\right) - \Gamma\frac{\boldsymbol{\omega}_0}{\omega_0}\left(\frac{\boldsymbol{\omega}_0\bullet\boldsymbol{\sigma}}{\omega_0} - \sigma^{\mathrm{eq}}\right),$$

Here Γ is the relaxation rate and

$$\sigma^{\mathrm{eq}} = \tanh\frac{\hbar\omega_0}{2k_BT}$$

is the equilibrium magnetization — **Becomes Curie-Weiss equation**

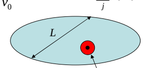
Dipolar field: $W = g\mu_B S(B_z + B_{i,z}^{(D)}) = W_{\mathrm{ext}} + W_i^{(D)}$

where

$$W_i^{(D)} = E_D D_{i,zz}, \qquad E_D = \frac{(g\mu_B S)^2}{v_0}, \qquad D_{i,zz} = \sum_j \phi_{ij}\sigma_{jz}$$

and

$$\phi_{ij} = v_0\frac{3(\mathbf{e}_z\bullet\mathbf{n}_{ij})^2 - 1}{r_{ij}^3}, \qquad \mathbf{n}_{ij} = \frac{\mathbf{r}_{ij}}{r_{ij}}$$



Uniformly magnetized ellipsoid: $D_{zz} = \sigma_z\sum_j\phi_{ij} \equiv \bar{D}_{zz}\sigma_z \qquad v_0^{1/3} \ll r \ll L$

Shape dependence $\bar{D}_{zz}^{(\mathrm{cyl})} = \bar{D}_{zz}^{(\mathrm{sph})} + 4\pi\nu(1/3 - n_z)$

Small sphere:
Inside summation
outside integration

n_z – demagnetizing factor
ν – number of sublattices, 2 for Mn12

$$\bar{D}_{zz}^{(\mathrm{sph})} = \begin{cases} 0, & \text{simple cubic} \\ 2.155, & \text{Mn}_{12} \text{ (body centered tetragonal)} \\ 4.072, & \text{Fe}_8 \end{cases} \Longrightarrow \bar{D}_{zz}^{(\mathrm{cyl})} = 10.53$$

Magnetic ordering

For $\Delta \ll E_D\bar{D}_{zz}$ the Curie-Weiss equation reads

$$\sigma_z(z) = \tanh\left(\frac{E_D}{2k_BT}D_{zz}(z)\right)$$

Uniform solution: $D_{zz} = \bar{D}_{zz}\sigma_z \Longrightarrow T_C = E_D\bar{D}_{zz}/k_B$

For Mn_{12} $E_D/k_B \simeq 0.0671$ K

Thus for a Mn_{12} cylinder: $T_C \simeq 0.782$ K

comparable with the experimental value 0.9 K, F. Luis et al, PRL-2005

Static domain wall

1*d* approximation

Inhomogeneously magnetized long cylinder of radius R:

$$D_{zz}(z) = \nu\int_{-L/2}^{L/2} dz' \frac{2\pi R^2\sigma_z(z')}{\left[(z'-z)^2 + R^2\right]^{3/2}} - k\sigma_z(z)$$

Local term

where

$$k \equiv 8\pi\nu/3 - \bar{D}_{zz}^{(\mathrm{sph})} = 4\pi\nu - \bar{D}_{zz}^{(\mathrm{cyl})} > 0,$$

$$k = 14.6 \text{ for Mn}_{12} \text{ and } k = 4.31 \text{ for Fe}_8$$

the Curie-Weiss equation

$$\sigma_z(z) = \tanh\left(\frac{E_D}{2k_BT}D_{zz}(z)\right)$$

is an integral equation!

σ_x is due to Δ and very small, the DW is linear (Ising-like)

Numerical solution for the DW profile

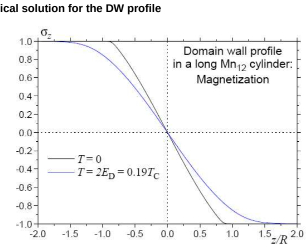


FIG. 1: Magnetization profile of a domain-wall in a Mn_{12} cylinder at two different temperatures.

Domain-wall mobility

For small B_z the DW speed v_{DW} is linear in B_z

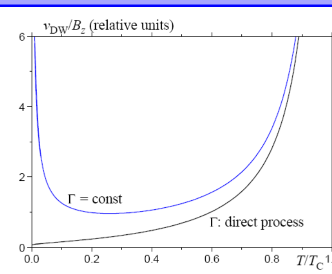
$$v_{DW} \equiv \mu_{DW}B_z, \qquad \mu_{DW} \propto \begin{cases} 1/\Gamma, & \text{standard DWs with } |\mathbf{m}| = \text{const} \\ \Gamma, & |\mathbf{m}| \neq \text{const (linear DWs)} \end{cases}$$

DW mobility μ_{DW} follows from the static DW profile from the energy balance:

$$v_{DW} = \frac{S\sigma_\infty g\mu_B B_z}{k_BT}\left[\int_{-\infty}^{\infty} dz \frac{1}{\Gamma}\frac{1}{1-\sigma_z^2}\left(\frac{d\sigma_z}{dz}\right)^2\right]^{-1}$$

Direct phonon processes:

$$\Gamma = \frac{S^2\Delta^2\omega_0\left(g\mu_B H_\perp\right)^2}{12\pi E_t^4}\coth\frac{\hbar\omega_0}{2k_BT}$$



At, e.g., $S = 10$, $B_z = 0.1$ T, $T = 1$ K, and $R = 1$ mm, this gives $v_{DW} \sim 1$ m/s for $\langle\Gamma\rangle = 10^3$ s^{-1} and $v_{DW} \sim 10^3$ m/s for $\langle\Gamma\rangle = 10^6$ s^{-1}.

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