SPIN SEMICLASSICS

ANUPAM GARG
(NORTHWESTERN UNIVERSITY)

BOULDER SUMMER SCHOOL 2003
FRONTIERS OF MAGNETISM

MOTIVATION

- MENTAL PICTURES OF QM OFTEN CLASSICALLY BASED. SC LIMIT IMPORTANT IN "UNDERSTANDING" QTM. MECH.
- QUANTITATIVELY ACCURATE IN MANY CASES:

  LINEARIZED SPIN WAVES (ANDERSON, 1950's)
  1-D SPIN CHAINS (HALDANE, 1983; 1985)

- MOST VISUAL APPROACH — PATH INTEGRALS

  "STRINGING PHASES ONTO CLASSICAL TRAJECTORIES"

- PARTICLES $\left[\mathcal{H} = \frac{1}{2m}p^2 + V(x)\right]$
  - FEYNMAN PATH INTEGRAL $\left[\ \langle x_f | e^{-i\mathcal{H}t} | x_i \rangle\ \right]$
  - SC LIMIT WELL UNDERSTOOD

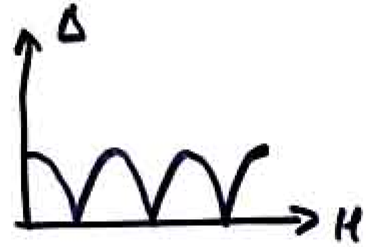
    VAN VLECK (1928)
    ⋮ MASLOV PHASE; FEYNMAN-SOIREAM, ...
    GUTZWILLER (1970's)
    ⋮

- SPIN $\left[\ \mathcal{H} = fn.(J_x, J_y, J_z)\right]$
  - ONLY PATH INTEGRAL BASED ON COHERENT STATES
  - SC LIMIT

    KLAUDER (1979)
    SOLARI (1987)
    ⋮ A WORK IN PROGRESS.

TOPIC I: THE Fe_8 SYSTEM.

- SPIN ORIENTATION TUNNELING IN Fe_8
  - EXAMPLE WITH INTERESTING PHYSICS VIA SPIN PATH INTEGRAL.
  - RESONANCE LIKE FLIP-FLOP IN NH_3.
- INTERFERING SEMICLASSICAL PATHS
  - TUNNEL SPLITTING (Δ) OSCILLATES WITH H.



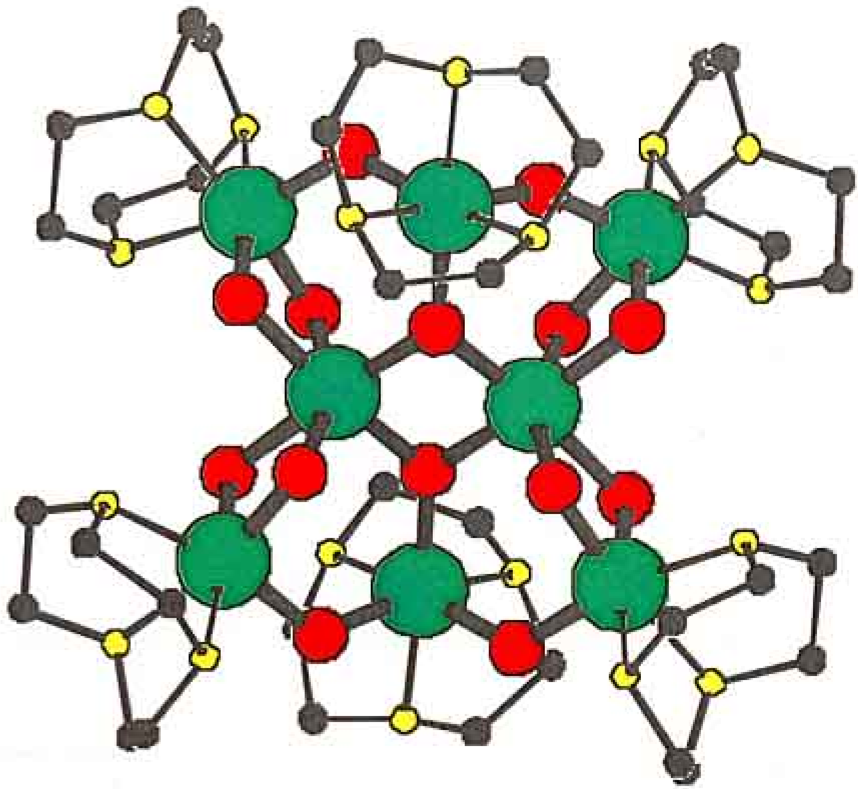
(DO NOT CONFUSE WITH THE FLIP-FLOP IN TIME.)

- NO. OF ZEROS IN Δ:
  - NAIVELY, "TOPOLOGICAL", = 10.
  - EXPERIMENTALLY, SEE 4.
- MISSING ZERO PROBLEM
  - INCLUDE DISCONTINUOUS PATHS!

TOPIC II: SPIN PATH INTEGRAL & SC LIMIT

- CLASSICAL DYNAMICS FOR SPIN (POISSON BRACKETS)
- QUANTIZE VIA PATH INTEGRAL
- LEAST ACTION/SEMICLASSICAL PATHS
- EXPLICIT BOUNDARY TERMS IN ACTION/HPF.
  - PATHS MUST BE COMPLEXIFIED
    $\hat{n}(t) \equiv \{\theta(t), \phi(t)\}$; θ, ϕ BOTH COMPLEX
  - SOLVES OVERDETERMINATION PROBLEM
  - YIELDS HAMILTON-JACOBI EQNS.
- GAUSSIAN FLUCTUATIONS
  - GLOBAL ANOMALY
  - STANDARD METHOD (JACOBI'S EQN.) FAILS
  - EXTRA TERM (SOLARI, 1987; KOCHETOV, 1995).
- DO FOR PARTICLE COHERENT STATE PATH INTEG.
  - SAME PROBLEM, SAME SOLN., SIMPLER.
- BOHR-SOMMERFELD QUANTIZATION RULE.

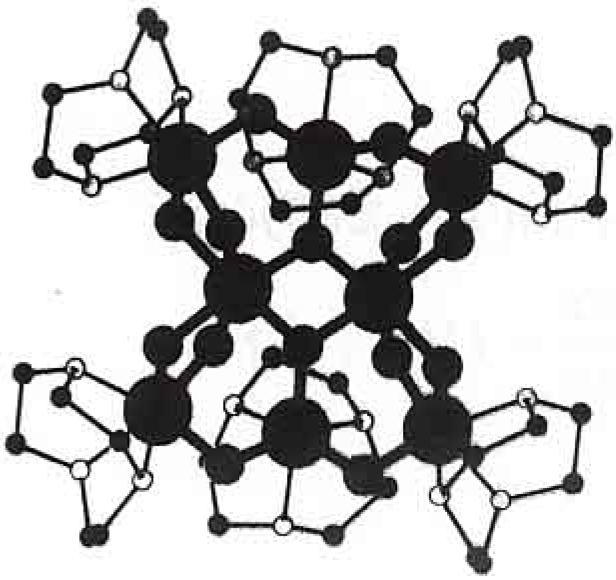
Magnetic molecular clusters Fe_8 with $S = 10$

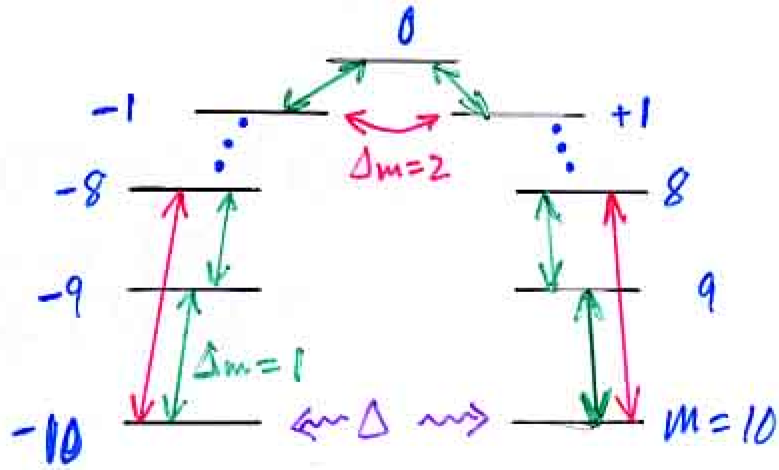


The Fe_8 System

$[Fe_8(tacn)_6O_2(OH)_{12}]^{8+}$

approximate D_2 symmetry, almost planar arrangement



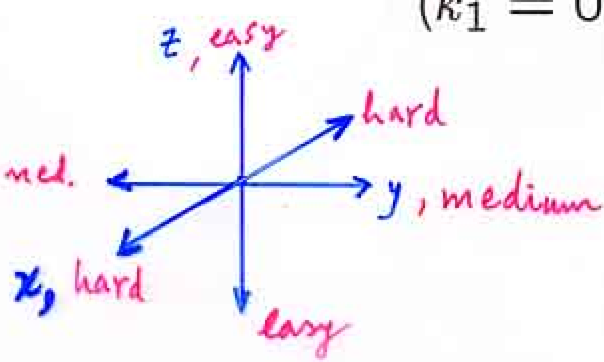


ferrimagnetic cluster ground state of S = 10

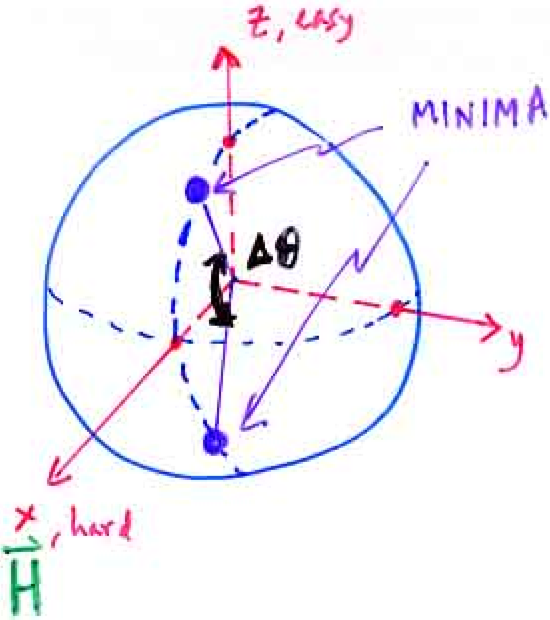
- spin $J = 10$, about 40 active e^- spins
- molecule crystallizes with different Fe_8's parallel
- Fe_8's far apart; no exchange; v. weak dipolar coupling $\Longrightarrow$ single molecule problem
- with extra dc field along $\hat{x}$ (hard axis)

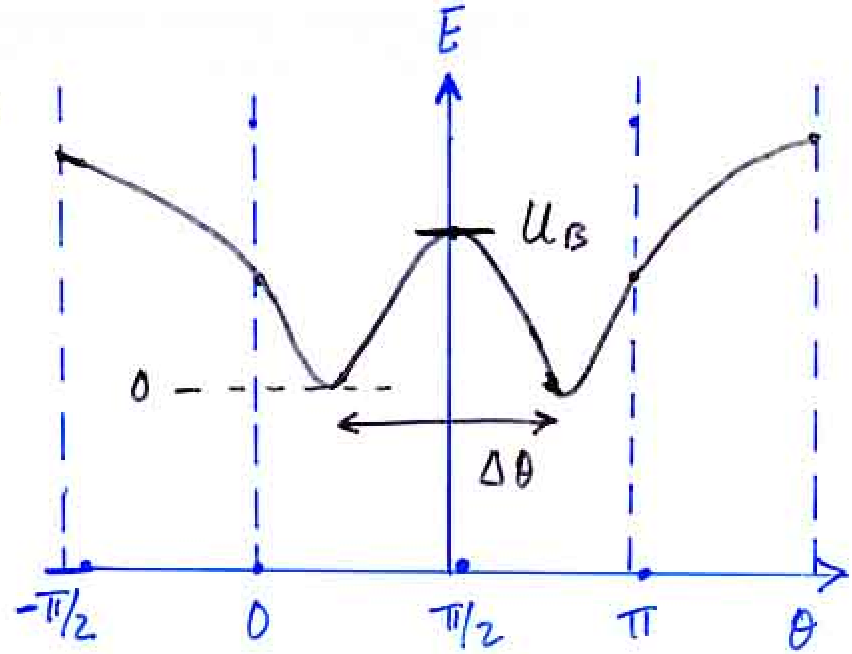
$$\boxed{\mathcal{H} = -k_2 J_z^2 + (k_1 - k_2) J_x^2 - \gamma H J_x}$$

$(k_1 = 0.33\ \text{K}, k_2 = 0.22\ \text{K})$

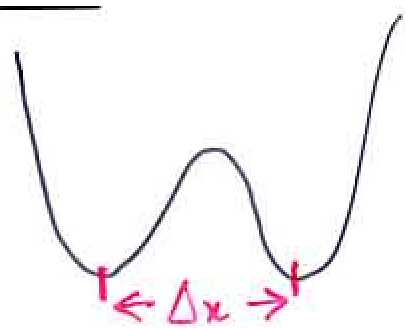


Δ vs. H_{HARD} – WHAT TO EXPECT





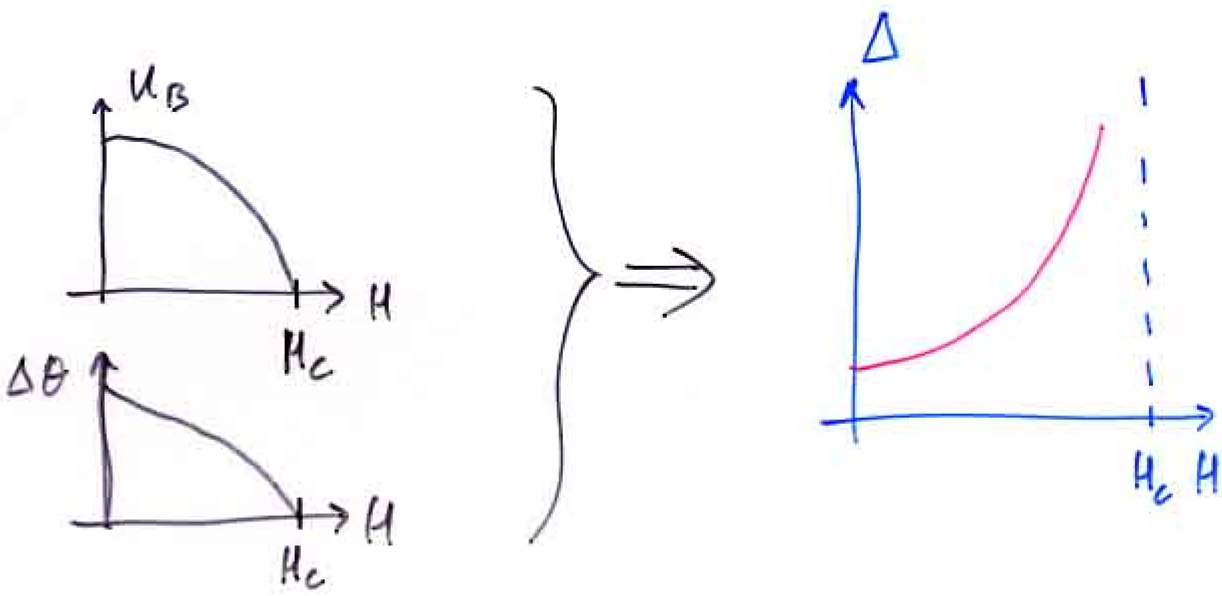
- 1d particle



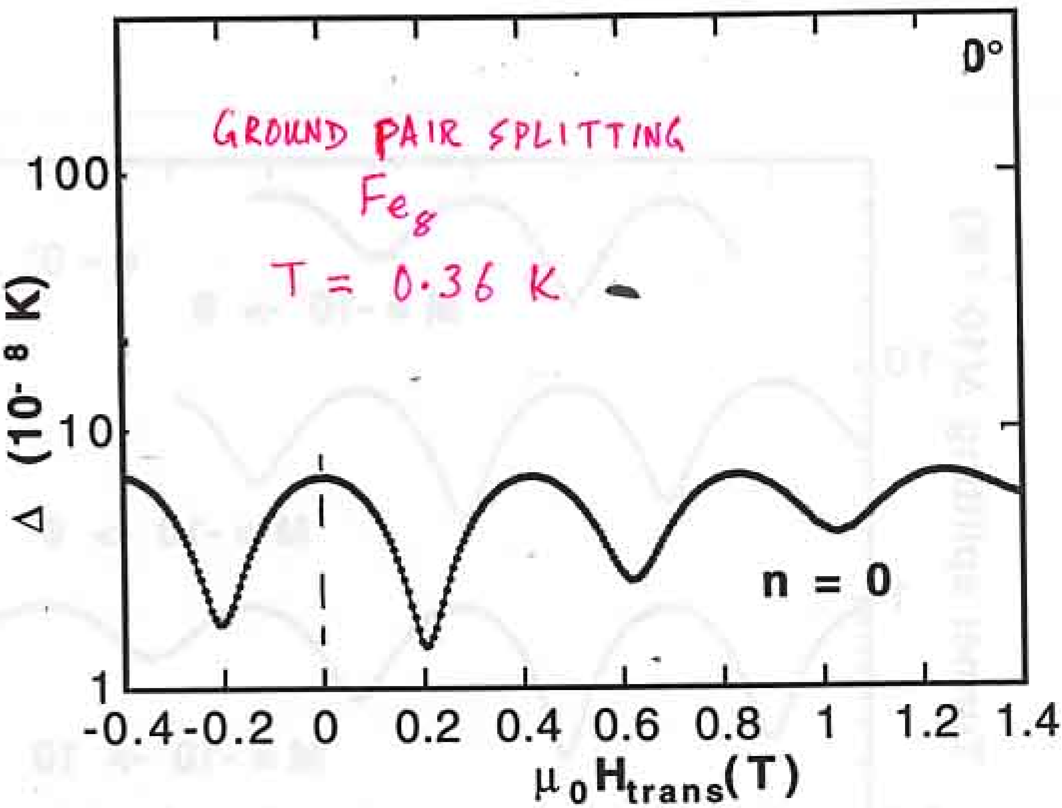
Barrier height or Δx ↘

Tunnel splitting ↗

- spin



Δ vs. H_{HARD} – WHAT IS SEEN

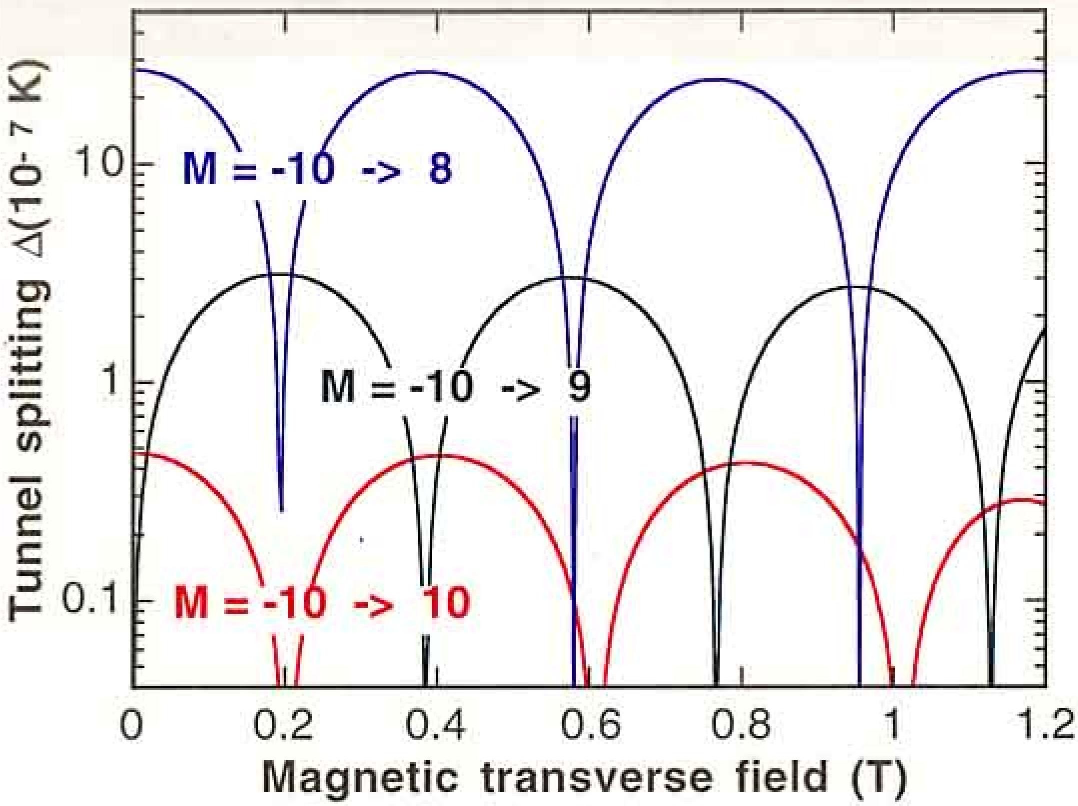


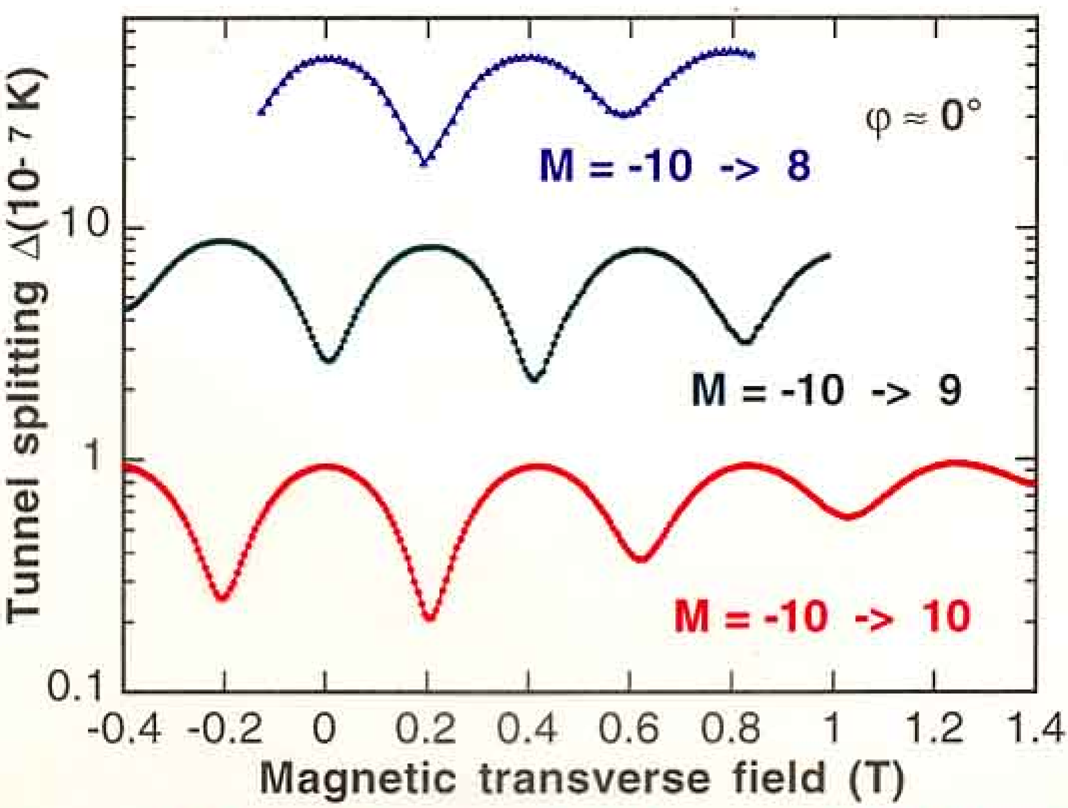
W. WERNSDORFER & R. SESSOLI

SCIENCE 284, 133 (1999).

(Similar effects now seen in $Mn_{12}^{[2-]}$.)

Parity effect of the tunnel splitting ocillations

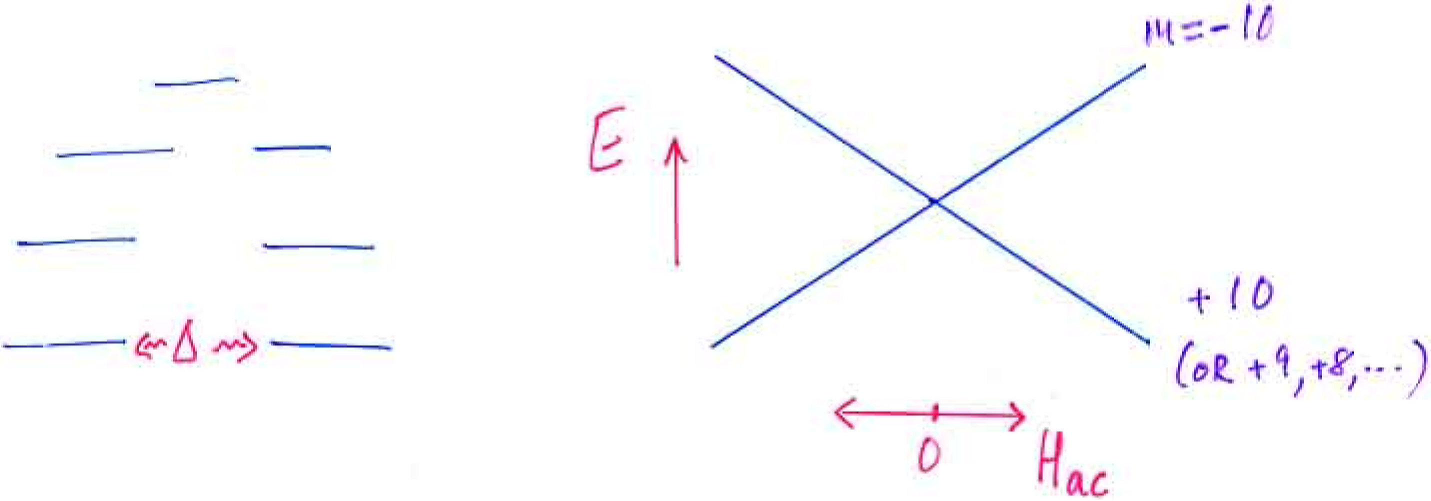




W. Wernsdorfer and R. Sessoli, Science, 284, 133 (1999).

Measurement of Δ

- Δ is very small ($\sim 10^{-8}$ K). How to measure?
- Apply small ac field along *easy axis* ($\hat{\mathbf{z}}$).



- Landau-Zener-Stückelberg process (1932)

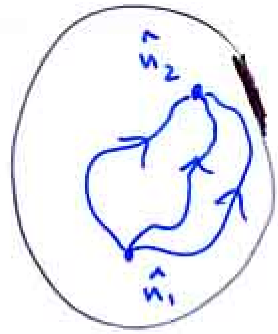
$$\Gamma_{\text{LZS}} \sim \frac{\Delta^2}{H_{\text{ac}}}$$

- Extract Δ from measurement of Γ_{LZS}.

INTERFERING TUNNELING PATHS

- STANDARD INSTANTON CALC$\underline{^N}$

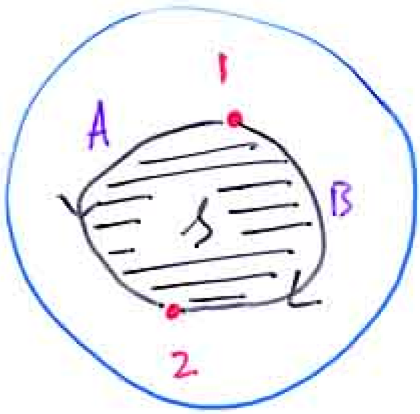
$$\langle \hat{n}_2 | e^{-HT} | \hat{n}_1 \rangle \sim \int_1^2 [d\hat{n}]\, e^{-S[\hat{n}]}$$



- USUALLY, ONE PATH DOMINATES — INSTANTON.

- T-REVERSAL (OR SOME OTHER SYMMETRY)

  $\Rightarrow$ TWO (OR MORE) EQUIV. INSTANTONS



$$\boxed{S_A - S_B = iJ\,\Omega(\mathcal{S})}$$

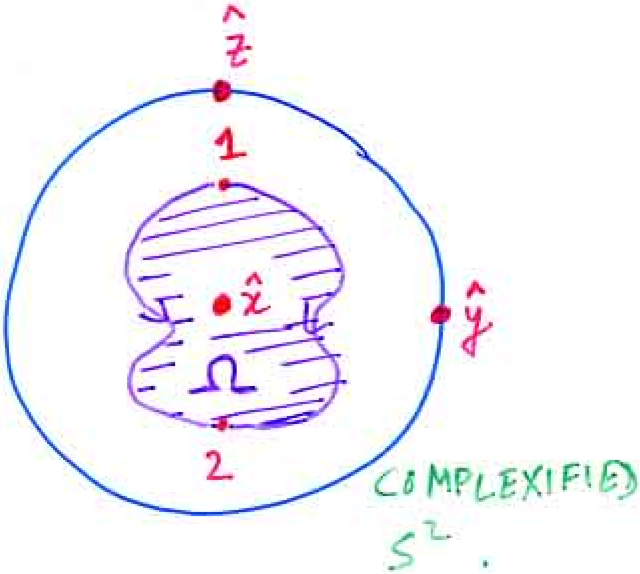
- Interference!
  spin-parity effects, Kramers,
  J dependence of tunneling.

- LOSS, DIVINCENZO & GRINSTEIN
- VON DELFT & HENLEY

$\Big\}$ PRL 1992

also, HALDANE GAP; 1983, 85.

Δ OSCILLATIONS IN Fe_8.

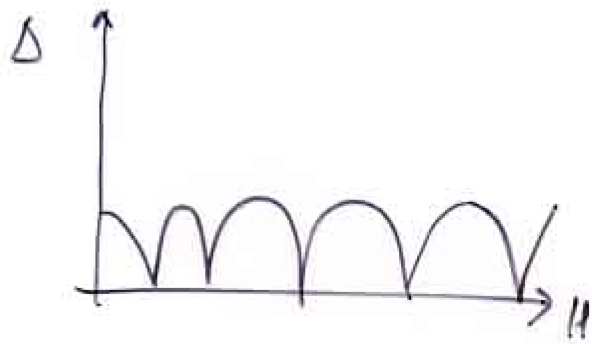


TWO EQUIVALENT INSTANTONS IF $\vec{H} \parallel$ HARD AXIS.

- $$\Delta \propto e^{-Re(S_{INST})}\left(1 + e^{iJ\Omega}\right)$$

  $$\propto (\#) \cos\left(\frac{J\Omega}{2}\right)$$

- Ω DEPENDS ON H.



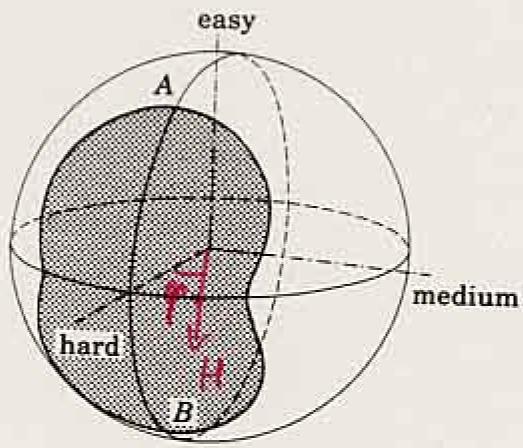


Fig. 1

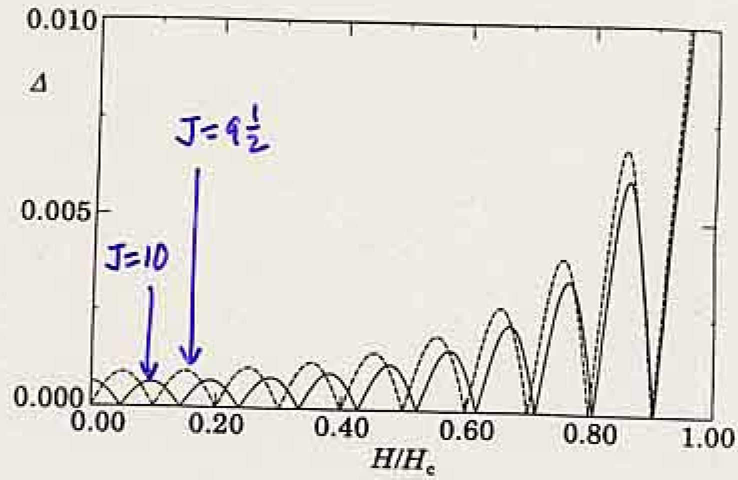
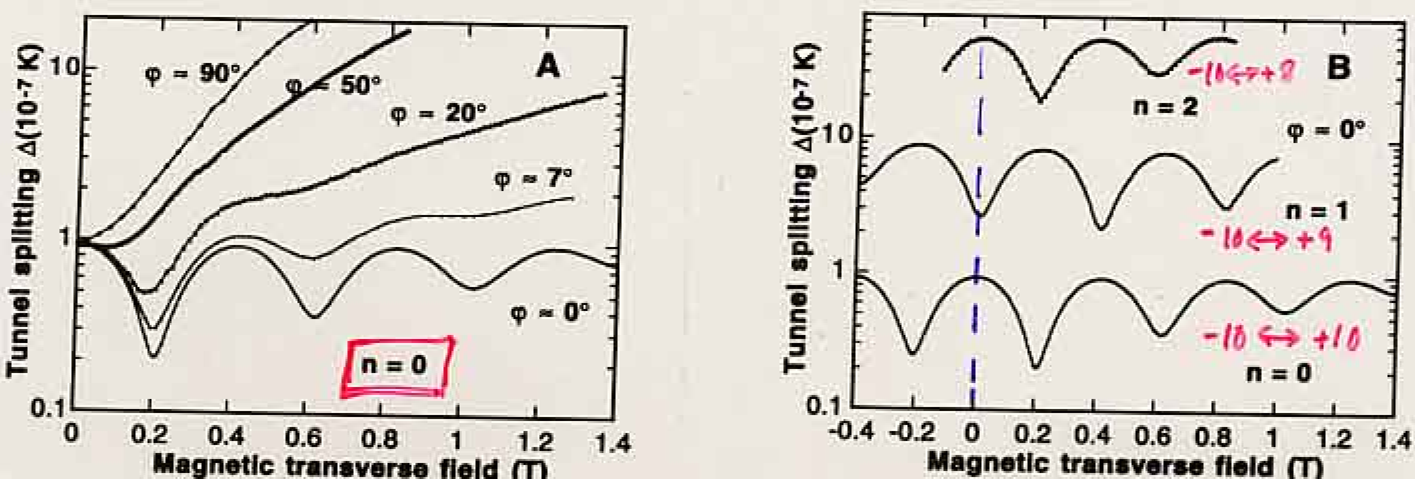


Fig. 2

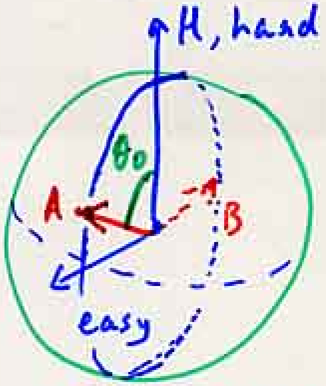
AG, EPL, 1993

$$\Delta \propto \cos\left(\frac{\pi}{2}\,\text{Area}\right)$$



Wernsdorfer & Sessoli, Science 284, 133 (1999).

INTERFERING INSTANTON CALC$^{\underline{N}}$



$$S_E = \int_{-T/2}^{T/2} d\tau \left[iJ(1-\cos\theta)\dot{\phi} + E(\theta,\phi) \right]$$

A&B: CLASSICAL MINIMA

$$E(\theta,\phi) = k_1 J^2 (\cos\theta - \cos\theta_0)^2 + k_2 J^2 \sin^2\theta \sin^2\phi$$

$\cos\theta_0 = H/H_c$.

$k_2/k_1 \equiv \lambda < 1$.

E-L EQN:

$$iJ \frac{d\hat{n}}{d\tau} = -\hat{n} \times \frac{\partial E}{\partial \hat{n}}$$

$$\therefore \quad dE/d\tau = 0.$$

SO, NEED NOT SOLVE FOR $\hat{n}(\tau)$ EXPLICITLY.

PUT $E(\theta,\phi) = E_{min} = 0$ & SOLVE FOR $\theta(\phi)$.

$$\boxed{S_{INST.} = iJ\int(1-\cos\theta)\,d\phi}$$

$$\cos\theta = \frac{\cos\theta_0 + i\sqrt{\lambda}\sin\phi\,(1-\cos^2\theta_0 - \lambda\sin^2\phi)^{1/2}}{(1-\lambda\sin^2\phi)}$$

$$\frac{1}{J}\operatorname{Im}(S_{INST}) = \int_0^{\pm\pi}\left(1 - \frac{\cos\theta_0}{1-\lambda\sin^2\phi}\right)d\phi = \pm\pi\left(1 - \frac{H/H_c}{\sqrt{1-\lambda}}\right)$$

- NOTE: PATHS RUN ON COMPLEX UNIT SPHERE.

INTERFERING INSTANTONS (CONTD.)

TUNNEL SPLITTING

$$\Delta \sim \sum_{\substack{\text{ALL} \\ \text{INSTANTONS}, j}} e^{-S_j^{\text{EUCLIDEAN}}}$$

HERE, $S_\pm = \mathrm{Re}(S) \pm i \, \mathrm{Im}\, S_\pm$

$$S_+ - S_- = i J \Omega$$

Area of loop on Complex sphere.

$$\Delta \simeq \left(e^{-\mathrm{Re}(S)}\right) \cos\left[\frac{J\Omega}{2}\right]$$

$$\Omega = \pi\left(1 - \frac{H/H_c}{\sqrt{1-\lambda}}\right)$$

$\Delta = 0$ if $J\Omega =$ odd integer $\times \pi$

i.e. $\frac{H}{H_c} = \sqrt{1-\lambda}\left(1 - \frac{n+1/2}{J}\right)$

(INTEGER J
$n = 0, 2, 3, \ldots, J-1 \Rightarrow J$ zeros for $H > 0$.

HALF-INT. J $\quad n = 0, 2, \ldots, J - \frac{1}{2} \Rightarrow J + \frac{1}{2}$ zeros for $H \geq 0$.)

OTHER SPIN/PSEUDO-SPIN MODELS

- LIPKIN-MESHKOV-GLICK (1965)

  - Collective excitations like giant resonances in nuclei

$$\mathcal{H} = \epsilon\left[J_z + \alpha\left(J_x^2 - J_y^2\right)\right]$$

- MOLECULAR ROTORS

e.g. SF_6 : $\mathcal{H} = \beta\left(J_x^4 + J_y^4 + J_z^4\right)$

  - Harter & Patterson 1977, 1984
  - Robbins, Creagh & Littlejohn 1989, 1990

- SUPERDEFORMED NUCLEAR ROTORS (^{149}Gd, ^{153}Dy, ^{184}Hg)

$$\mathcal{H} = A J_z^2 + B_1\left(J_x^2 - J_y^2\right)^2 + B_2\left(J_x^2 + J_y^2\right)^2$$

  - Hamamoto & Mottelson, 1994, 1995

- "ORDER FROM DISORDER" IN QTM. AFM'S

  - TUNNELLING & BS QUANTIZ'N OF SPIN CLUSTERS.

  - Henley & Zhang, 1998, 1999 ...

Lagrangian for spin

- $\mathcal{H} =$ function of $J_x, J_y, J_z \equiv U(\mathbf{J})$

$$\frac{d\mathbf{J}}{dt} = \{\mathbf{J}, \mathcal{H}\}_{\mathsf{PB}} = -\mathbf{J} \times \nabla_{\mathbf{J}} U$$

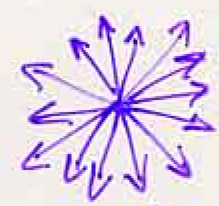
- Constants of motion: $\mathbf{J}\cdot\mathbf{J}, \quad U(\mathbf{J})$.

- Rewrite eqn. of motion like charged particle in $\mathbf{B}$-field. ($\mathbf{J}$ like position of particle on a sphere)

$$\begin{aligned} 0 &= \frac{d\mathbf{J}}{dt} \times \mathbf{J} + J^2 (\nabla_{\mathbf{J}} U)_{\perp \text{ to } \mathbf{J}} \\ \overset{0}{\cancel{m}}\ddot{\mathbf{x}} &= \mathbf{v} \times \mathbf{B} - \nabla V \end{aligned}$$

- $\mathbf{x} \leftrightarrow (\mathbf{J}/J) = \hat{\mathbf{n}}, \qquad \underbrace{\mathbf{B} \leftrightarrow \mathbf{J} = J\hat{\mathbf{n}}}$.

$$L = \frac{1}{2}\overset{0}{\cancel{m}}\dot{x}^2 + \mathbf{A} \cdot \frac{d\hat{\mathbf{n}}}{dt} + U$$



$\vec{B}_{pseudo}$

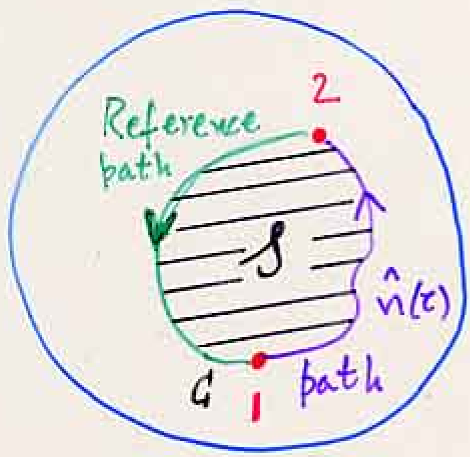
- Note: $\mathbf{A}$ is vector pot'l for monopole field.

Action for spin

- $L = \mathbf{A} \cdot \frac{d\hat{\mathbf{n}}}{dt} + U(\mathbf{J}), \quad (\hat{\mathbf{n}} = \frac{\mathbf{J}}{J}, \nabla \times \mathbf{A} = J\hat{\mathbf{n}})$

$$S = \int_{t_1}^{t_2} L\, dt = \int_1^2 \mathbf{A} \cdot d\hat{\mathbf{n}} + \int U\, dt$$

Unit Sphere



Ref. path like choice of gauge.

$$\begin{aligned} \int_1^2 \mathbf{A} \cdot d\hat{\mathbf{n}} \;\rightarrow\; & \int_{1\to 2} + \int_{\text{ref. path } 2\to 1} = \oint_C \mathbf{A} \cdot d\hat{\mathbf{n}} \\ = & \int_S (\nabla \times \mathbf{A}) \cdot \hat{\mathbf{n}}\, d^2 s \\ = & J\Omega(\mathcal{S}) \quad \boxed{(\Omega = \text{Area})} \end{aligned}$$

$$S_{\text{path}} = J\,\Omega(\mathcal{S}_{\text{path}}) + \int_{\text{path}} U\, dt$$

- $\Omega \to \Omega + 4\pi$; $e^{iJ\Omega}$ UNCHANGED.

THE MISSING QUENCH PROBLEM

- WS SEE ONLY 4 QUENCHES, NOT 10.

- MEASURED "PERIOD" = 0.41 T

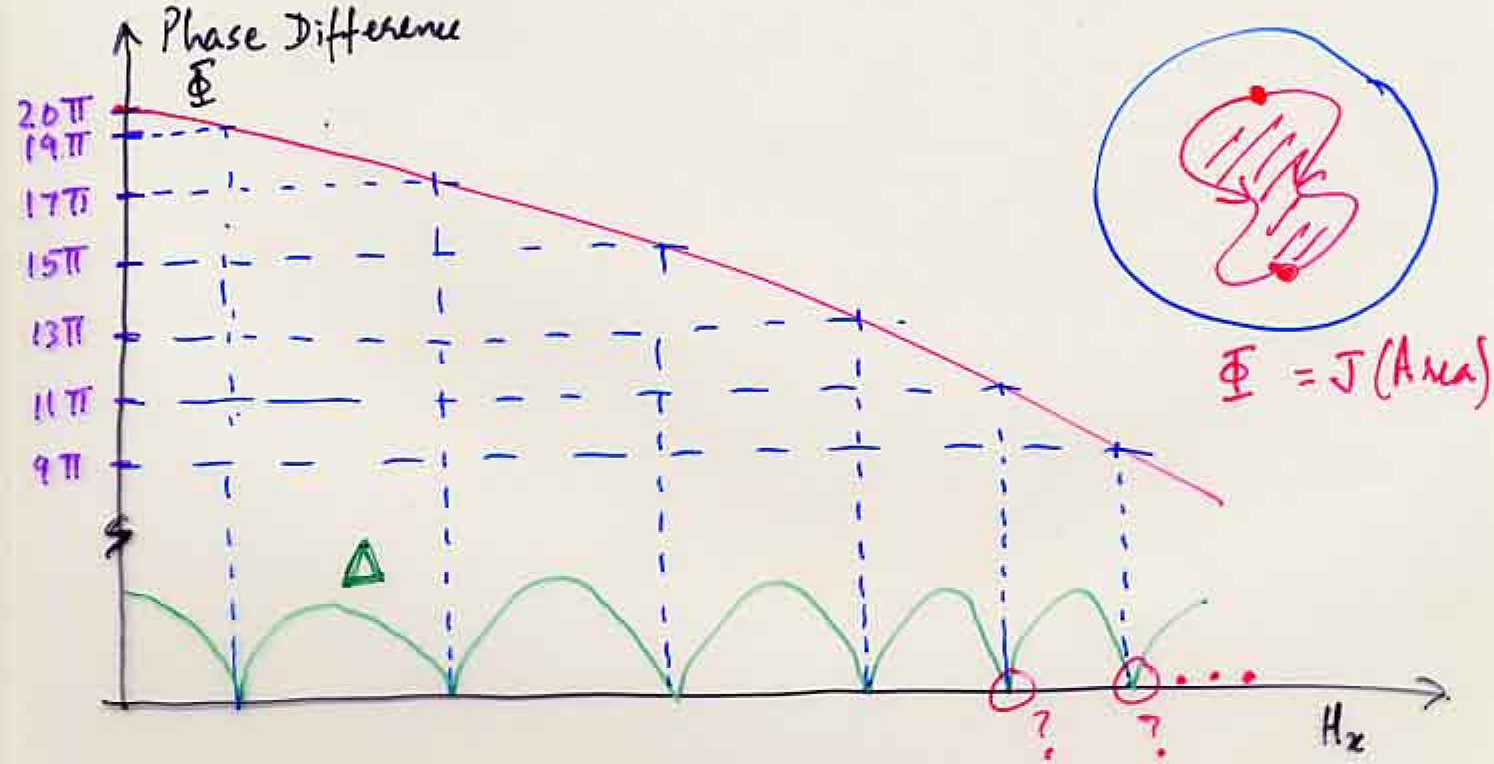
  THEORY (Garg 1993) $\rightarrow$ 0.26 T

- WS POSIT V. WEAK 4th ORDER ANISOTROPY

$$\mathcal{H} = k_1 J_x^2 + k_2 J_y^2 - C\left(J_+^4 + J_-^4\right)$$

$$C J^2 / k_1 \sim 10^{-3}$$

- NUMERICS CONFIRMS ONLY 4 QUENCHES.



DISCONTINUOUS INSTANTONS

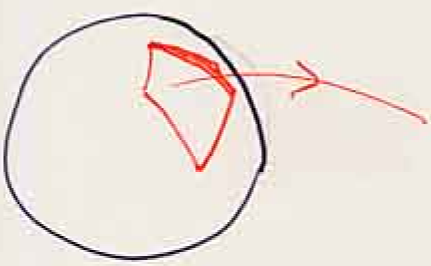
- SPIN TRAJECTORIES NEED NOT BE CONTINUOUS

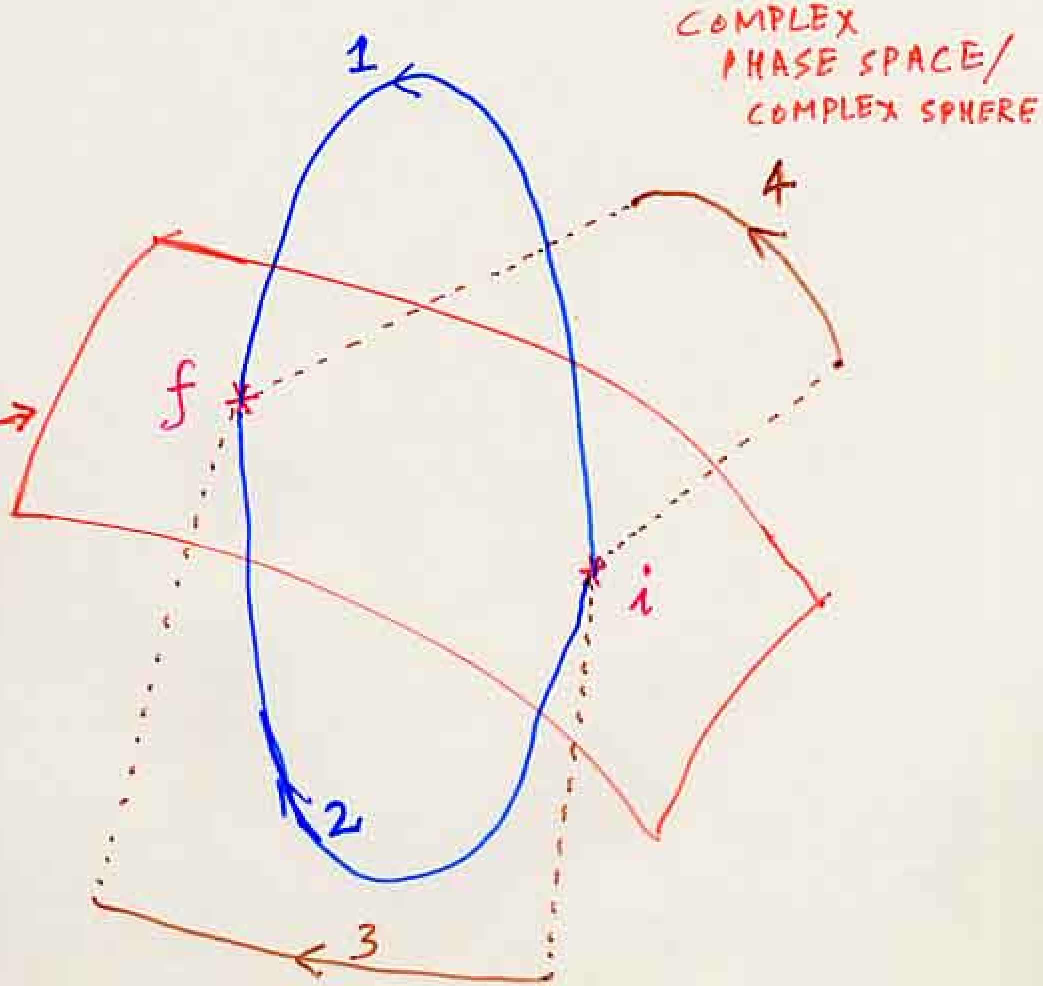
  – KLAUDER, 1979.

$|\theta,\phi\rangle$: SPIN-COHERENT STATE ALONG (θ,ϕ) DIRECTION.

$$\langle\theta,\phi\,|\,\theta',\phi'\rangle \neq 0\,!$$

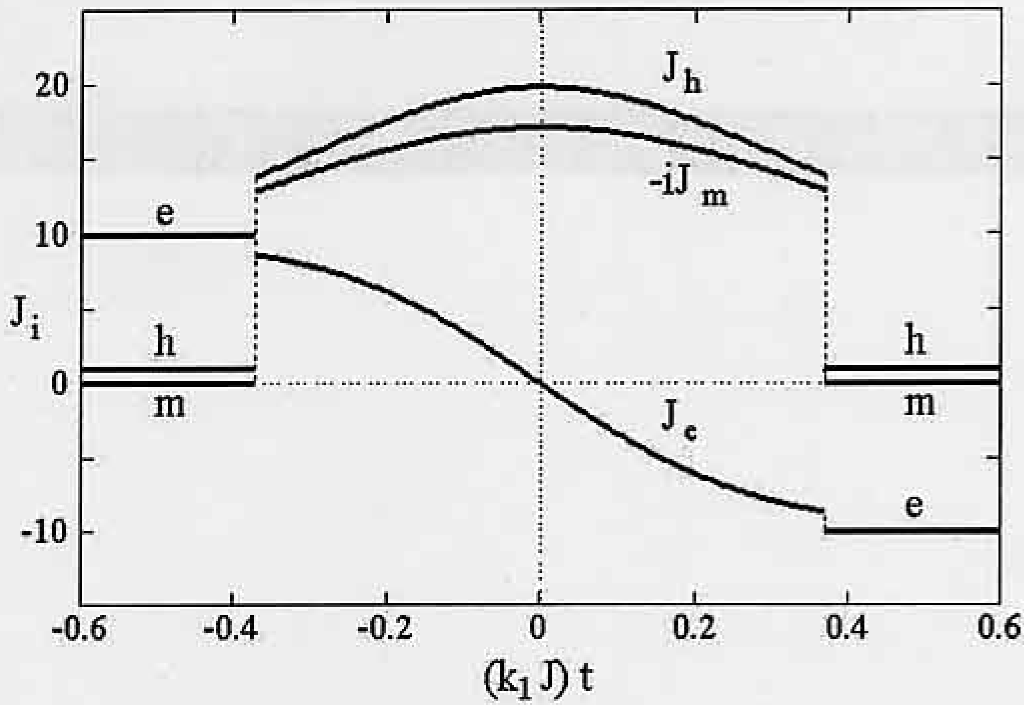
- 4 TH ORDER TERM IN Fe_8 INTRODUCES SUCH TRAJECTORIES AMONG THE CLASSICAL PATHS. (SINGULAR PERTURB$^{\underline{N}}$)



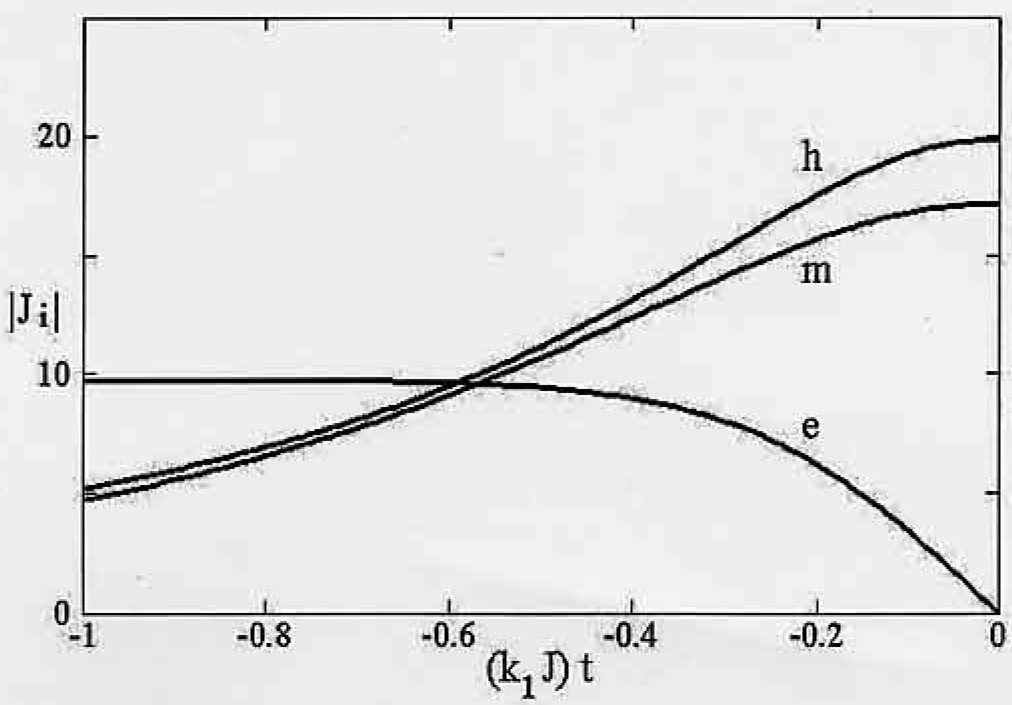


1 & 2 – Interfering Instantons

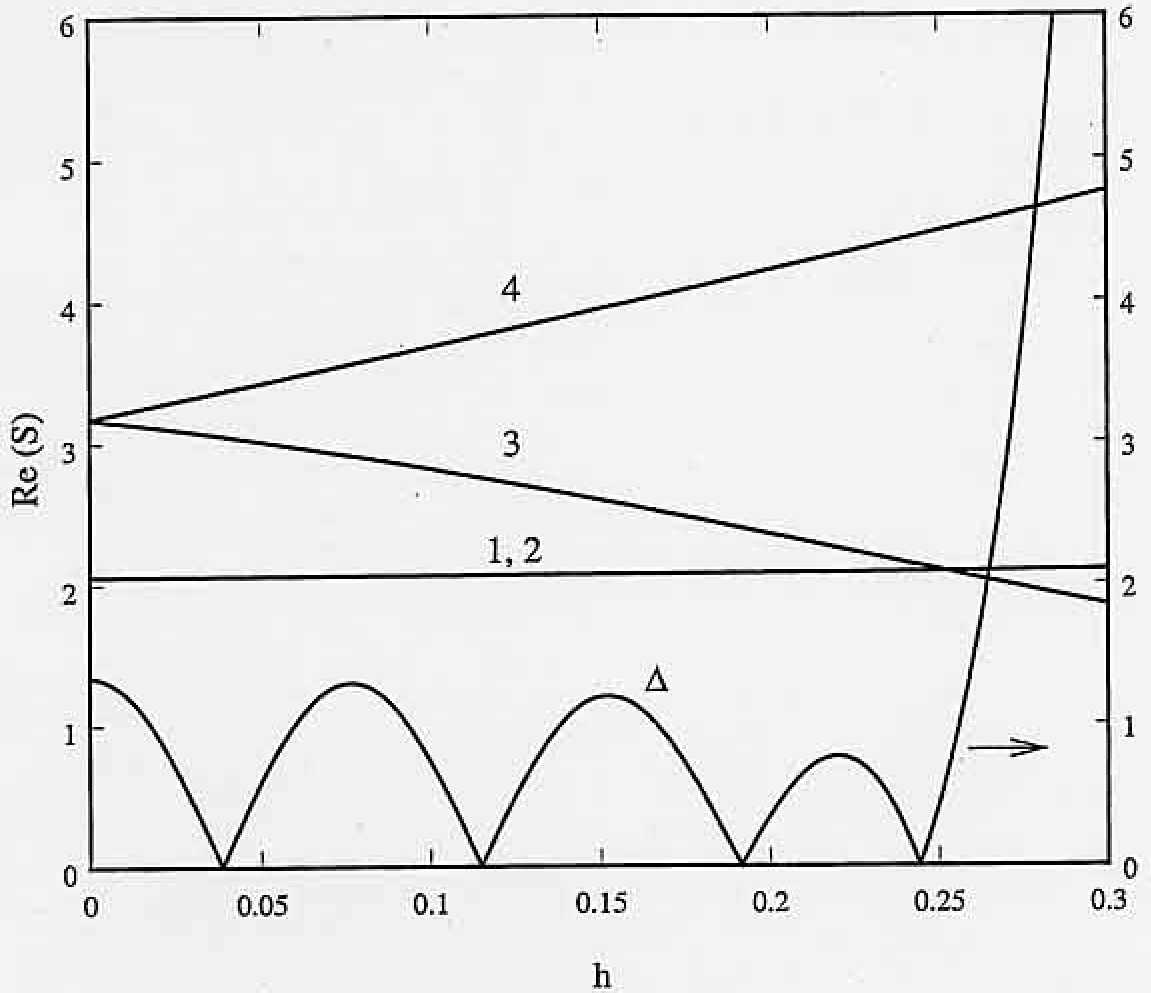
3 & 4 – Non interfering Instantons



Boundary Jump Instanton (#3)



Non-Jump Instantons (#1,2)



- INTERFERENCE SHUNTED BY DISCONTINUOS PATHS AT LARGE H.

E. Kececioglu & AG, PRL 88, 237205 (2002)
cond-mat/0207115
↳ PRB 67, 054406 (2003).

Notes on Lecture 1.

① There are several other approaches to the semiclassical limit for spin, especially for tunneling calculations. None of them has the geometrical appeal of the coherent state path integral, but they are still valuable.

(a) The earliest work is by
I. Ya. Korenblit & E.F. Shender, Sov. Phys. JETP 48 (1978).

(b) Map spins to particles (by Villain transformation e.g.) + path integrals:
M. Enz & R. Schilling, J. Phys. C 19, L711 & 1765 (1986).
V. I. Belinicher, C. Providencia, J. da Providencia, J. Phys. A: Math. Gen. 30, 5633 (1997).

(c) Write Schrödinger eqn. in J_z basis & solve recursion relation (discrete WKB / Phase integral method):
J.L. van Hemmen & A. Suto, EPL 1, 481 (1986); Physica 141B, 37 (1986).
J. Villain & A. Fort, Euro. Phys. J. B 17, 69 (2000).
AG, PRL 83, 4385 (1999); math-ph/0003005.

(d) The idea of using instantons based on the coherent-state action is due to
E. Chudnovsky & L. Gunther, PRL 60, 661 (1988).

(e) The so-called longitudinal basis is discussed nicely in
S. Takagi, Macroscopic Qtm. Tunneling, Camb. U. Press (2002).

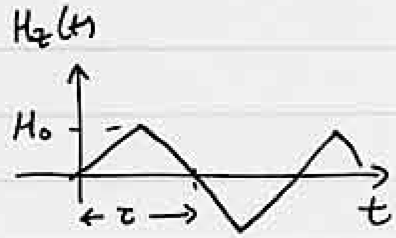
---

② The LZS process was independently discussed in the same year by
(a) L.D. Landau, Phys. Z. Sowjetunion 2, 46 (1932).
(b) C. Zener, Proc. Roy. Soc. London A 137, 696 (1932).
(c) E.C.G. Stuckelberg, Helv. Phys. Acta 5, 369 (1932).

Zener's paper is very clear. See also the section on "predissociation" in Landau & Lifshitz, Qtm. Mech. 3rd ed, Sec. 90.

For the present problem, one gets

$$\Gamma_{LZS} = \frac{\pi \quad \Delta^2}{(g\mu_B \hbar)(\Delta m)\; H_0}.$$



H_0 = amplitude of a <u>triangular</u> wave

(Δm) = change in m qtm. no. due to tunneling.

The result for Γ_{LZS} is independent of τ; this is seen exp'tly for $|\dot{H}_z|$ ranging from $1\,mT/s$ to $1\,T/s$.

---

③ The instanton method is lucidly explained in

(a) S. Coleman, Aspects of Symmetry (Camb. U. Press, 1985), Chap 7.

(b) It was discovered by J. S. Langer, Ann. Phys. (N.Y.) <u>41</u>, 108 (1967), but Coleman's exposition is superior. See also

(c) S. Coleman, Phys. Rev. D <u>15</u>, 2929 (1977).

---

④ The integrals for the real part of the action for the simple model for Fe_8 are elementary. See,

AG, Phys. Rev. B <u>60</u>, 6705 (1999)

for this, and for what happens at fields beyond the 10th quench.