Low Dimensional Magnetism Workshop

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Pietro Gambardella

ICREA and Centre d'Investigació en Nanociència i Nanotecnologia (ICN-CSIC), Barcelona, Spain

Olivier Fruchart Institut Néel (CNRS), Grenoble, France

Wulf Wulfhekel Physikalisches Institut, Universität Karlsruhe, Karlsruhe, Germany



Outline

1. P. Gambardella (P. F. de Châtel)

- Magnetic order in one- and two-dimensional systems
- Influence of magnetic anisotropy on magnetic order

2. P. Gambardella

- Intrinsic magnetization properties and dimensionality effects: magnetization, orbital moments, induced magnetism in nonmagnetic materials
- Magnetocrystalline anisotropy
- Metal-organic layers and single-molecule magnets

3 O. Fruchart

- Superparamagnetism
- Thermal activation, nucleation and propagation of domains
- Magnetization reversal in thin films and nanoparticles

4. W.Wulfhekel

• Magnetic excitations in thin films and clusters probed by STM

Prototype 1D and 2D crystals



Cs⁺(((CH₃)₄N)⁺)
 Ni²⁺(Mn²⁺)

• F⁻(C{/⁻)

TMMC tetra methyl ammonium manganese chloride 1D, FM interaction

Reviews:

- Hone and Richards, Annu. Rev. Mater. Sci. 4, 337 (1974).
- de Jongh and Miedema, Adv. Phys. 50, 947 (2001 publ. 1974).
- Steiner, Villain, and Windsor, Adv. Phys. 25, 87 (1976).



Low-dimensional magnetism in metal nanostructures



Magnetic domains in 2D films









particles

3D or 2D

magnetic

1D nanowires

0D impurities



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Magnetic order in low-dimensional systems

Ferromagnetic

Localized Models: Heisenberg, Ising, ...

point-like interacting magnetic moments

- X Non-integer magnetic moments in metals
- \checkmark Modelization of thermal effects (spin waves), interpretation of T_c
- Modelization of spatial effects (magnetic domains)

Itinerant Models:

Based on electronic band structure, delocalized electrons

- Non-integer magnetic moments in metals
- Prediction of ferromagnetism in Fe, Co, Ni; nonmagnetic 3d, 4d, 5d metals
 - Temperature dependence of M
- Spatial dependence of M
 - (e.g., due to magnetostatic interactions in homogeneous materials)

see also the lecture by E. Burzo

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Magnetic coupling between atoms: inter-atomic exchange

• There are many possible exchange-interaction Hamiltonians, e.g.:

$$H = -J_{z} \sum_{i \neq j} S_{i}^{z} S_{j}^{z} - J_{\perp} \sum_{i \neq j} \left(S_{i}^{x} S_{j}^{x} + S_{i}^{y} S_{j}^{y} \right) \quad \begin{array}{l} \textit{Anisotropic Heisenberg} \\ \textit{model and XY model (J_{z}=0)} \end{array}$$

$$H = -J_z \sum_{i \neq j} \mathbf{S}_i^{\mathbf{z}} \mathbf{S}_j^{\mathbf{z}}$$

Ising model

Lattice binding energy + magnetic energy

Moruzzi et al., Handbook of Mag. Materials Vol. 7

Heisenberg Hamiltonian in the presence of an external field

Need for approximations: e.g., nearest neighbor interaction, mean-field

See lecture by C. Lacroix

$$\mathbf{S}_{i} = \left(\mathbf{S}_{i} - \langle \mathbf{S}_{i} \rangle\right) + \left\langle \mathbf{S}_{i} \rangle$$

$$\mathbf{S}_{i} \cdot \mathbf{S}_{j} = \left(\mathbf{S}_{i} - \langle \mathbf{S}_{i} \rangle\right) \cdot \left(\mathbf{S}_{j} - \langle \mathbf{S}_{j} \rangle\right) + \left(\mathbf{S}_{i} - \langle \mathbf{S}_{i} \rangle\right) \cdot \left\langle \mathbf{S}_{j} \right\rangle + \left(\mathbf{S}_{j} - \left\langle \mathbf{S}_{j} \right\rangle\right) \cdot \left\langle \mathbf{S}_{i} \right\rangle + \left\langle \mathbf{S}_{i} \right\rangle \cdot \left\langle \mathbf{S}_{j} \right\rangle$$
Fluctuations
(correlated deviations
from thermal average)
$$\mathbf{0}$$

$$\Rightarrow \mathbf{S}_{i} \cdot \mathbf{S}_{j} \approx 2\mathbf{S}_{i} \cdot \left\langle \mathbf{S}_{j} \right\rangle - \left\langle \mathbf{S}_{i} \right\rangle \cdot \left\langle \mathbf{S}_{j} \right\rangle$$

define
$$m(T) \coloneqq \frac{M(T)}{M(\theta)} = \frac{\left\langle S_i^z \right\rangle}{S}$$

$$H = -\frac{J}{2} \sum_{i,j} \mathbf{S}_{i} \cdot \mathbf{S}_{j} - g\mu_{B} \mathbf{B} \sum_{i} S_{i}^{z} \longrightarrow H \approx -(Jzm + g\mu_{B}B) \sum_{i} S_{i}^{z} + \frac{J}{2} Nzm^{2}$$

Number of neighbors
analogy with
paramagnetic case $\mathbf{m} = \mathfrak{B}_{S} \left(\frac{\mu_{B}gSB}{kT} \right) \longrightarrow \mathbf{m} = \mathfrak{B}_{S} \left(\frac{Jzm + \mu_{B}gSB}{kT} \right)$

 $T/T_c = 0.5$ 1.0 r FM $T/T_c=1$ 0.8 0.6 $T/T_c=2$ 0.4 ΡM 0.2 0 1.0 0.2 0.4 0.8 1.2 0.6 0 $m \longrightarrow$

Effective field (Weiss molecular field)

Graphical solution: intersection of $\mathfrak{B}_{s}\left(\frac{Jzm}{kT}\right)vs.m$ with line representing m For small arguments $\mathfrak{B}_{s}(x) \approx \frac{S+I}{3S}x$

Limit intersection gives T_c

 $T_C = \frac{S+1}{3S} \frac{J_{exc} z}{k}$

Temperature dependence of the magnetization in the FM Heisenberg model

 $B_{\text{Weiss}} = \frac{Jz \,\mathrm{m}}{g \,\mu_{R} S}$

The Heisenberg model provides an atomistic explanation of the Weiss field for a FM material

The Curie temperature depends on the number $T_{C} = \frac{S+1}{3S} \frac{J_{exc}z}{k}$ of nearest neighbors z. E.g., for bcc Fe, z = 8, T_{c} = 1040 K, S ≈ 2, we get $J \approx 20$ meV.

 $\Rightarrow T_c$ nanostructures < T_c bulk

Experimental temperature dependence of the magnetization in thin films

 $T_C = \frac{S+1}{3S} \frac{J_{exc}Z}{k} \quad \Longrightarrow$

1/t dependence

Critical Temperatures of Ising Lattice Films

G. A. T. ALLAN Baker Laboratory, Cornell University, Ithaca, New York 14850

Phys. Rev. B 1, 352 (1970)

 $T_C(\infty) - T_C(t) \sim t^{-\lambda}, \quad \lambda = 1$

Gradmann, Handbook of Mag. Materials Vol. 7

This model accounts for the decrease of T_c with *t* down to a critical thickness $t_0 \approx 4$ monolayers. Below t_0 ferromagnetic order can become unstable ($T_c = 0$) depending on the magnetic anisotropy.

$$M(T) = M(0) \left(1 - \frac{T}{T_c}\right)^{\beta}$$

$$\mathbf{m} \approx \left(T_C - T\right)^{\beta}$$

Mean Field	3D Ising	2D Ising	1D Ising
$\beta = 1/2$	$\beta = 0.33$	$\beta = 1/8$	-

FIG. 2. Critical exponent β as a function of film thickness (error bars are given in Table I). The dashed lines show the theoretical values for a 3D Heisenberg, 3D Ising, and 2D Ising system. The shaded regime marks the crossover from 3D to 2D.

For a review of critical parameters in magnetic materials see, e.g., Kadanoff Rev. Mod. Phys. 39, 395 (1967).

Limitations of Mean Field theory:

• It does not take into account spin waves and fluctuations

• T_C depends on the number of neighbors only, but in reality also on the lattice dimensionality and symmetry.

Excitated states of the Heisenberg Hamiltonian correspond to magnons

$$H = -\frac{J}{2} \sum_{i,j} \mathbf{S}_{i} \cdot \mathbf{S}_{j}$$
Excited states, $T > 0$

$$|\mathbf{R}_{i}\rangle = \frac{1}{\sqrt{2S}} \mathbf{S}_{-}(\mathbf{R}_{i})|0\rangle = |\mathbf{S}\rangle_{\mathbf{R}_{i}} |\mathbf{S}\rangle_{\mathbf{R}_{2}} \dots |\mathbf{S}\rangle_{\mathbf{R}_{N}}$$

$$|\mathbf{q}\rangle = \sum_{\mathbf{R}_{i}} e^{i\mathbf{q}\cdot\mathbf{R}_{i}} |\mathbf{R}_{i}\rangle$$

$$H = \mathbf{E}_{0} - \sum_{\mathbf{q}} \hbar \omega(\mathbf{q}) \mathbf{a}_{\mathbf{q}}^{\dagger} \mathbf{a}_{\mathbf{q}}$$

$$\hbar \omega(\mathbf{q}) \sim 2JSa^{2}q^{2} = \mathbf{D}q^{2}$$
spin wave stiffness

R

Hamiltonian
$$H = E_0 - \sum_q \hbar \omega(\mathbf{q}) a_q^{\dagger} a_q$$

Magnetization
at finite T $M(T) = M(0) - \mu_B S \frac{1}{V} \sum_q \langle n_q \rangle$, $\langle n_q \rangle = \frac{1}{e^{\hbar \omega(q)/kT} - 1}$
 $\Rightarrow M(0) - M(T) \Rightarrow \mu_B S \frac{1}{(2\pi)^3} \int_{B.Z.} \frac{d\mathbf{q}}{e^{\hbar \omega(q)/kT} - 1} \approx \mu_B S \frac{1}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{d\mathbf{q}}{e^{\ln^2/kT} - 1}$
In 3D, Bloch $T^{3/2}$ dependence:
Substitution and
integration on solid angle $\frac{Dq^2}{kT} \Rightarrow x \Rightarrow M(0) - M(T) \approx \mu_B S \frac{1}{4\pi^2} \left(\frac{kT}{D}\right)^{3/2} \int_0^{\infty} \frac{\sqrt{x}}{e^x - 1} dx$
Small wavevector, finite T approximation:
 $\int_{-\infty}^{\infty} \frac{d\mathbf{q}}{e^{\ln^2/kT} - 1} \approx \int_{-\infty}^{\infty} \frac{d\mathbf{q}}{1 + D\mathbf{q}^2/kT - 1} \propto \int_{-\infty}^{\infty} \frac{d\mathbf{q}}{\mathbf{q}^2}$ Diverges in less than 3D
Absence of magnetic order in 2D and 1D at $T > 0$

F. Bloch, Z. Phys. 61, 206 (1930); N.D. Mermin and H. Wagner, Phys. Rev. Lett. 17, 1183 (1966).

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PHYSICAL REVIEW LETTERS

ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin[†] and H. Wagner[‡]

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York (Received 17 October 1966)

It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin-S Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.

VOLUME 87, NUMBER 13 PHYSICAL REVIEW LETTERS

24 September 2001

Absence of Spontaneous Magnetic Order at Nonzero Temperature in One- and Two-Dimensional Heisenberg and XY Systems with Long-Range Interactions

P. Bruno*

Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany[†] (Received 6 May 2001; published 7 September 2001)

The Mermin-Wagner theorem is strengthened so as to rule out magnetic long-range order at T > 0in one- or two-dimensional Heisenberg and XY systems with *long-range* interactions decreasing as $R^{-\alpha}$ with a sufficiently large exponent α . For *oscillatory* interactions, ferromagnetic long-range order at T > 0 is ruled out if $\alpha \ge 1(D = 1)$ or $\alpha > 5/2(D = 2)$. For systems with *monotonically decreasing* interactions, ferro- or antiferromagnetic long-range order at T > 0 is ruled out if $\alpha \ge 2D$.

0

(a)

 $H_{k}^{eff} > 0$

Heisenberg model: low-T spin wave excitations -> collapse of magnetic order in 2D and 1D Model assumes ideal isotropic spins

In a real magnetic material, the presence of an effective magnetic anisotropy field

opens a gap at the bottom of the spin-wave excitation spectrum of width

Ë

 $\Delta = 2\mu_B H_K^{eff} \quad \text{Out-of-plane} \qquad \Delta' = 2\mu_B \frac{H_{dip}}{8} \sqrt{\frac{2\mu_B H_K^{eff}}{2JS}} \quad \text{In-plane}$

0

(b)

 $H_{\kappa}^{eff} < 0$

E

The presence of magnetic anisotropy allows for ferromagnetic order to set in

 \Rightarrow finite CurieTemperature

$$T_C = \frac{4\pi(S+1)}{3} \frac{2JS}{\ln(8\pi JS/\Delta)}$$

N.B. 1: T_C depends logarithmically on the magnetocrystalline (out-of-plane case) or dipolar (in-plane case) anisotropy, but linearly on the number of n.n. and exchange. T_C is only weakly influenced by the magnetic anisotropy.

N.B. 2:
$$\Delta > \Delta' \implies T_C^{\perp} > T_C^{\parallel}$$

P. Bruno, Phys. Rev. B 43, 6015 (1991);
P. Bruno, Mater. Res. Soc. Symp. Proc. 231, 299 (1992);
M. Bander and D. L. Mills, Phys. Rev. B 38, 12015 (1988).

NxN localized moments

exact solution (due to Onsager) : long-range magnetic order prevails for
$$kT < \frac{2J}{\ln(1+\sqrt{2})}$$

A 2D Ising system is less sensitive to thermal fluctuations with respect to the 1D case. A finite Curie teperature exists, given by

$$T_c = \frac{2J}{k\ln\left(1+\sqrt{2}\right)}$$

"DEAD" LAYERS IN FERROMAGNETIC TRANSITION METALS*

L. Liebermann and J. Clinton University of California, La Jolla, California 92037

and

D. M. Edwards and J. Mathon[†] Imperial College, London, S. W. 7, England (Received 1 June 1970)

"Magnetically "dead" layers [...] are observed in nickel. From the temperature dependence of this effect it is deduced that two dead layers persist at T = 0, independent on the film thickness. [...] The existence of dead layers at T = 0 is attributed to a transfer of electrons from the s band to the d band in the neighborhood of a surface."

FIG. 1. Recording of the saturation flux as the thickness of a nickel film is continuously increased by electrolytic deposition. Four dead layers (or their equivalent) remain dead as the film thickness continues to grow.

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 $P = P_{Ni} (1 - e^{(x - x_0)/t})$

 $P_{Ni} \approx 15\%$ $x_0 \approx 1.2 \pm 1 \text{ \AA}$ 15 MAY 1974

Hot-electron scattering length by measurement of spin polarization

D. T. Pierce and H. C. Siegmann

Laboratorium für Festkörperphysik, Eidgenossische Technische Hochschule, CH-8049 Zürich, Switzerland

[...] A nonzero electron spin-polarization is observed in very thin Ni films showing that ferromagnetism occurs already in films of one or two layers average thickness.

FIG. 1. Polarization P as a function of Ni film thickness is given by the rectangular fields where the vertical dimension represents the statistical uncertainty (one standard deviation) and the horizontal dimension represents the estimated uncertainty of the average film thickness. Fields with the same cross hatching are for films successively evaporated on the same Cu substrate. The solid curve is a least-squares fit of an exponential (see text) to the points.

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Ferromagnetic Order in a Fe(110) Monolayer on W(110) by Mössbauer Spectroscopy

M. Przybylski^(a) and U. Gradmann

Physikalisches Institut, Technische Universität Clausthal, D-3392 Clausthal-Zellerfeld, Federal Republic of Germany (Received 22 June 1987)

"As strong crystalline anisotropies are present in monolayers, the physical problem is magnetic order in an anisotropic monolayer, not in the isotropic one."

FIG. 3. Magnetic hyperfine fields $B_{hf}(T)$ vs temperature for pseudomorphic Fe(110) monolayers on W(110). Mean values B_{hf} (filled circles) and B_{hf} for the magnetic component (open circles) for the Ag-coated layer [W(110)/Fe(0.82 ML)/Ag]. B_{hf} from extrapolation and $T_{C}(1)$ from thermal scan for the uncoated monolayer (circled crosses).

Experiments: magnetic order is present in 2D monolayer systems

1 ML Fe/Au(100), Cu(100) - S.D. Bader and E.R. Moog, J. Appl. Phys. 61, 3729 (1987) 1 ML Fe/Au(111) with $T_c \approx 315$ K - W. Dürr et al., Phys. Rev. Lett. 62, 206 - 209 (1989) 1 ML Co/Cu(100) with $T_c \approx 50$ K - C.M. Schneider et al., Phys. Rev. Lett. 64, 1059 (1990).

• Heisenberg model: at T > 0, no FM state in 1D and 2D

Mermin & Wagner, PRL 1966; Bruno, PRL 2001.

1D

- Anisotropic Heisenberg model and Ising model: FM state in 2D not in 1D

Ising, Z. Phys. 1925; Bander & Mills, PRB 1989

1D

ISING-LIKE MODEL

• Finite system (*N* localized moments):

ground state: $E=E_0$ lowest excited state: $E=E_0 + 2J$ (*N*-1 such states)

change in free energy: $\Delta F = \Delta E - T \Delta S = 2 J - kT \ln(N-1)$

 $\Delta F < 0 \Rightarrow$ no ferromagnetism for N > exp(2J/kT)

Interatomic exchange energy $2J \approx 15 \text{ meV}$

→ ferromagnetism is allowed only for

N < 50 atoms at T = 50 K

P. Gambardella et al., Phys. Rev. B 61, 2254 (2000); Surf. Sci. 449, 93 (2000)

P. Gambardella, J. Phys.: Condens. Matter 15, S2533 (2003).

Ferromagnetic order in 1D atomic chains: experiment

Monatomic Co chains

Magnetization curve: segments of about 15 atoms are ferromagnetically coupled

→ short-range FM order

Easy/hard axis magnetization: strong magnetic anisotropy (2.1 meV/atom)

Metastable FM state stabilized by large anisotropy barriers

long-range FM order

Nature 416, 301 (2002).

$$H = -\sum_{i=1}^{N-1} J \vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{S}}_{i+1} - \sum_{i=1}^{N} \left[K \left(\mathbf{S}_{i}^{z} \right)^{2} + g_{Co-Pt} \mu_{B} \vec{B} \cdot \vec{\mathbf{S}}_{i} \right]$$
$$M^{\alpha} = \frac{g_{Co-Pt}}{N} \sum_{i=1}^{N} \left\langle \mathbf{S}_{i}^{\alpha} \right\rangle$$

fixed parameters: N = 80 T = 45 K $g_{Co-Pt} = 3.8$, $||S_i|| = 1$

fitting parameters: J = 20 meV K = 3.3 meV

A. Vindigni, A. Rettori, M.G. Pini, C. Carbone, P. Gambardella, Appl. Phys. A 82, 385 (2006).

Spin-spin correlation matrix in zero field

A. Vindigni, A. Rettori, M.G. Pini, C. Carbone, P. Gambardella, Appl. Phys. A 82, 385 (2006).

Influence of magnetic anisotropy on the FM properties of 1D atomic chains

P. Gambardella et al., Phys. Rev. Lett. 93, 077203 (2004).

Coercive field and magnetization reversal in 1D atomic chains

Decrease of T_N with decreasing AFM thickness

FIG. 2. Temperature dependence of dc susceptibility at H = 100 Oe of representative multilayer samples of CoO/SiO₂ with a fixed SiO₂ layer thickness of 50 Å and various CoO layer thicknesses of t = 21, 25, 30, 34, and 39 Å. For clarity, the results are normalized to the maximum susceptibility.

CoO - T. Ambrose and C. L. Chien, Phys. Rev. Lett. 76, 1743 (1996).

FeF₂ - D. Lederman, C. A. Ramos, V. Jaccarino, and J. L. Cardy, Phys. Rev. B 48, 8365 (1993)

Usually, but not always, $T_B < T_N$, see Nogues et al., Phys. Rep. 422, 65 (2005) and refs. therein.

 $T_{\rm B}$ vs AFM film thickness in polycrystalline materials

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Existence of Phase Transitions for Anisotropic Heisenberg Models

Jürg Fröhlich*

Department of Mathematics, Princeton University, Princeton, New Jersey 08540

and

Elliott H. Lieb[†]

Department of Mathematics and Department of Physics, Princeton University, Princeton, New Jersey 08540 (Received 21 December 1976)

The two-dimensional anisotropic, nearest-neighbor Heisenberg model on a square lattice, both quantum and classical, has been shown rigorously to have a phase transition in the sense that the spontaneous magnetization is positive at low temperatures. This is so for all anisotropies. An analogous result (staggered polarization) holds for the antiferromagnet in the classical case; in the quantum case it holds if the anisotropy is large enough (depending on the single-site spin). *i.e.*, *if* $|S| \ge 1$

$$H = J \sum_{i,j} S_i \cdot S_j - K \sum_i \left(S_i^z \right)^2$$
$$= J \sum_{i,j} S_i^z S_j^z + J \sum_{i,j} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) - K \sum_i \left(S_i^z \right)^2$$

If J < 0 (FM) this term is zero since $|S_i| = \max$ ar favored

If J > 0 (AFM) this term mixes different spin configurations Hence the classical AFM ordered state is not an eigenstate of the Hamiltonian

S. Heinze et al., Science 288, 1808 (2000)

iii iii

AFM order in one-dimensional chains

a AFM alignment of classical spins in nanochains

b AFM quantum chains

P. Gambardella, Nat. Mater. 5, 432 (2006)

Inelastic spin-flip spectroscopy performed by STM reveals excited states of AFM

Heinrich et al., Science 306, 466 (2004), Science 312, 1021 (2006).

SUMMARY 1.

Magnetic order in low-dimensional metal systems

- > Spin lattice models
- Mean field approximation
- Mermin-Wagner theorem
- > Role of magnetic anisotropy
- > Experimental observation of magnetic order in 2D films and 1D chains
- Magnetic order in AFMs
- Experimental observation of AFM ground state in 2D and 1D systems