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Title: Bose-Einstein Condensation and Bose Glasses in an S = 1 Organo-metallic quantum magnet

Author(s): Zapf, Vivien

Intended for: University visit to the University of Waterloo

Abstract

I will speak about Bose-Einstein condensation (BEC) in quantum magnets, in particular the compound NiCl2-4SC(NH2)2. Here a magnetic field-induced quantum phase transition to XY antiferromagnetism can be mapped onto BEC of the spins. The tuning parameter for BEC transition is the magnetic field rather than the temperature. Some interesting phenomena arise, for example the fact that the mass of the bosons that condense can be strongly renormalized by quantum fluctuations. I will discuss the utility of this mapping for both understanding the nature of the quantum magnetism and testing the thermodynamic limit of Bose-Einstein Condensation.

Furthermore we can dope the system in a clean and controlled way to create the long sought-after Bose Glass transition, which is the bosonic analogy of Anderson localization. I will present experiments and simulations showing evidence for a new scaling exponent, which finally makes contact between theory and experiments. Thus we take a small step towards the difficult problem of understanding the effect of disorder on bosonic wave functions.

Bose-Einstein Condensation and Bose Glass in an S = 1 Organic Quantum Magnet

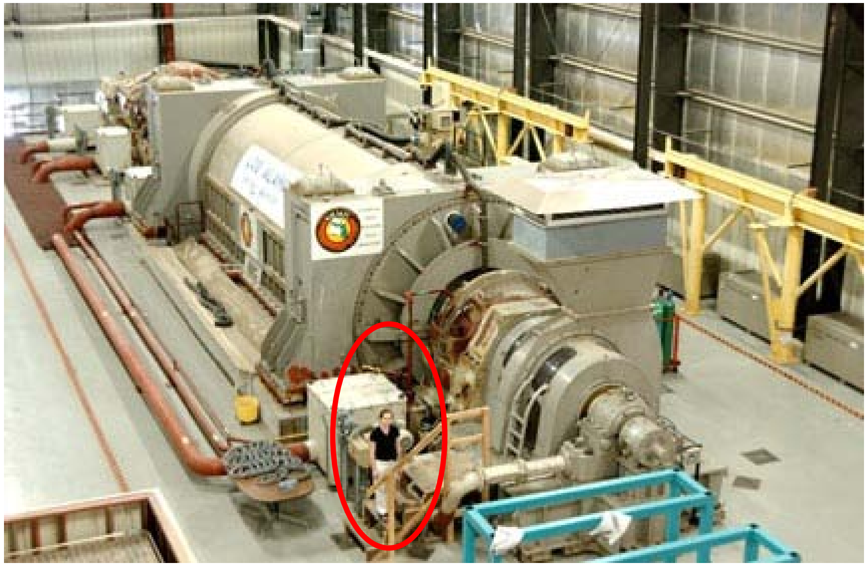
Vivien Zapf
National High Magnetic Field Laboratory
Los Alamos National Lab

Outline

- Overview of the magnet lab
- An experimentalist’s introduction to BEC in quantum magnets
- Experimental evidence for BEC in a Ni-based quantum magnet
- Bose Glass – BEC transition

National High Magnetic Field Laboratory – Pulsed Field Facility

Los Alamos National Lab



A user facility

•65 T pulses (milliseconds)

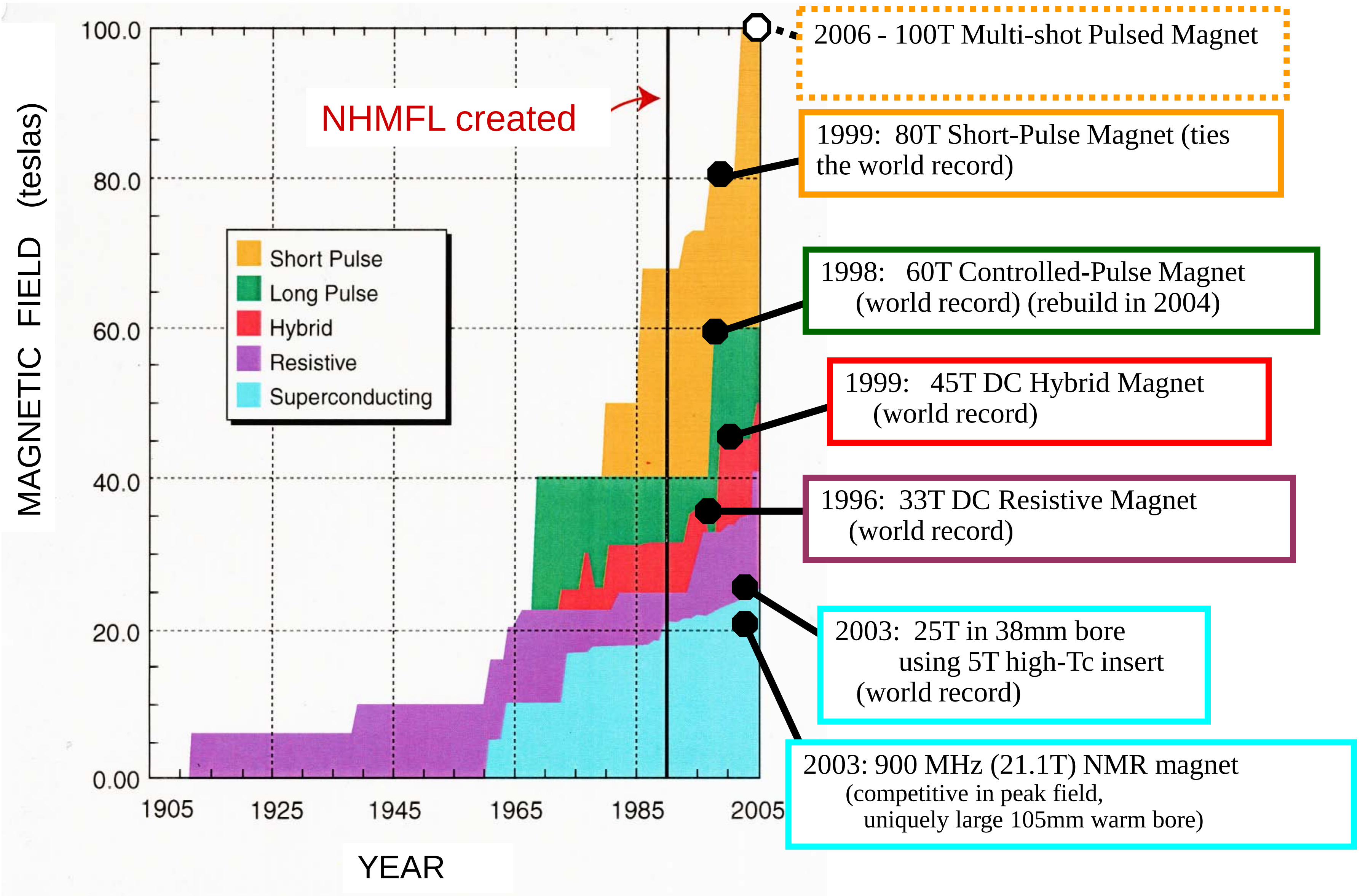
•60 T long pulse
(2 second shaped wave form)

•97.4 T (millisecond pulses)

•Thermodynamics, optics, electric, magnetic properties, electric polarization

•Accepting proposals! **magnet.fsu.edu**

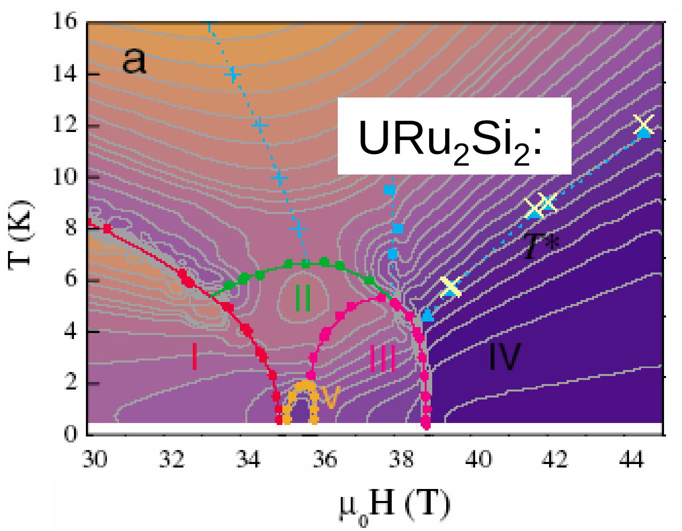
100 YEARS OF NON-DESTRUCTIVE MAGNETS



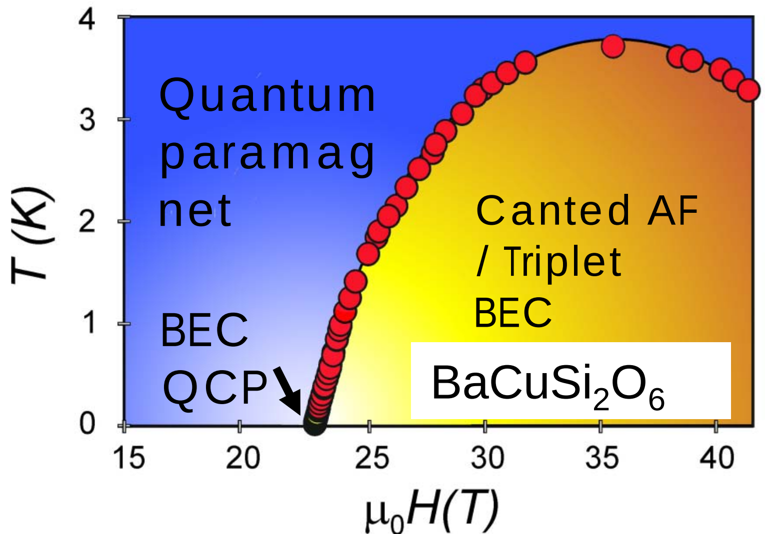
Why high magnetic fields?

- Induce new states of matter

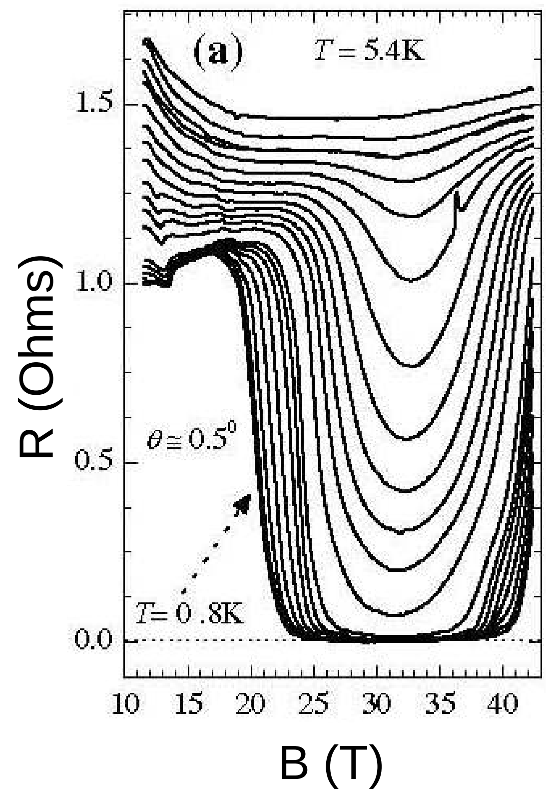
"Hidden order" state



Bose-Einstein Condensation



Field-induced superconductivity

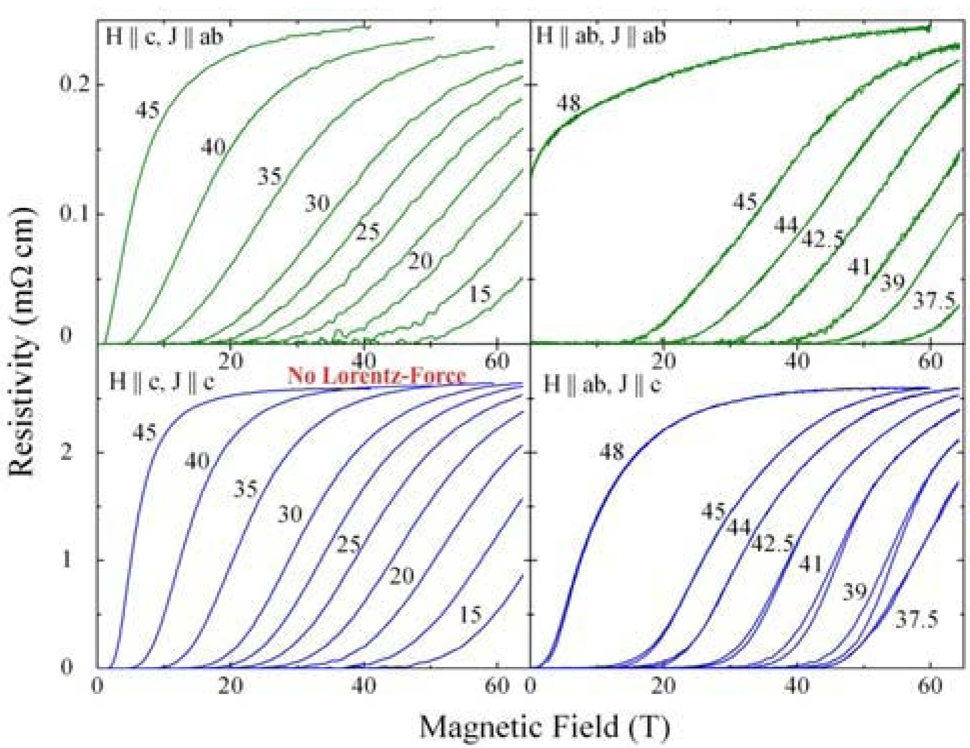


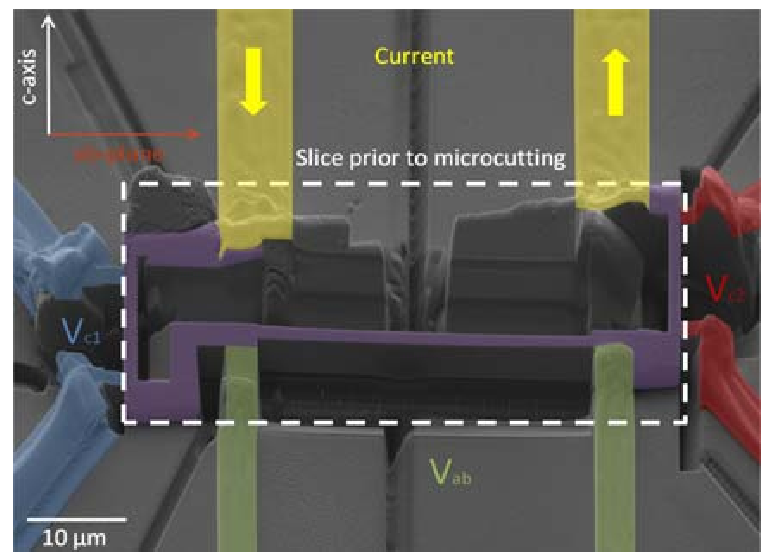
λ-$(BETS)_2$-$FeCl_4$

Why high magnetic fields?

- Induce new states of matter
- Destroy states of matter

(Understanding superconductivity by killing it…)

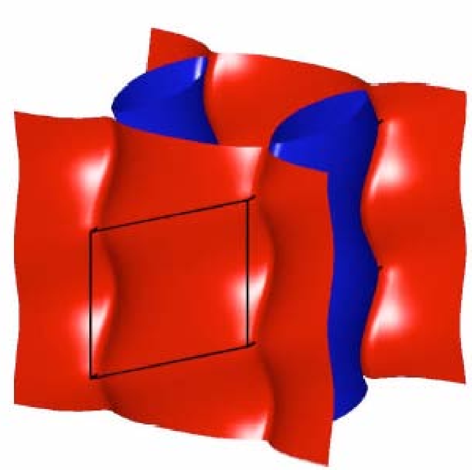




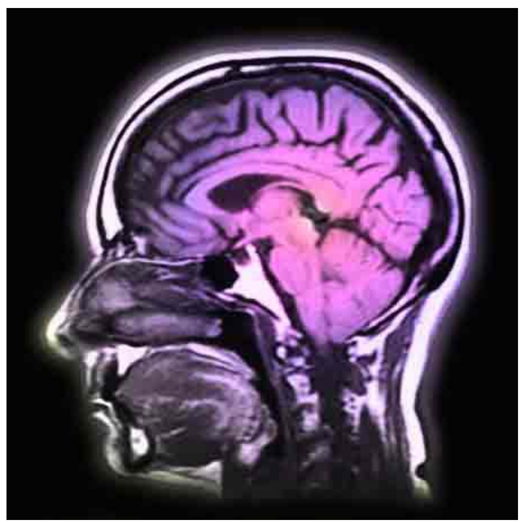
High magnetic-field scales and critical currents in SmFeAs(O, F) crystals
P. J. W. Moll, R. Puzniak, F. Balakirev, K. Rogacki, J. Karpinski, N. D. Zhigadlo, B. Batlogg,
Nature Materials 9, 628–633 (2010)

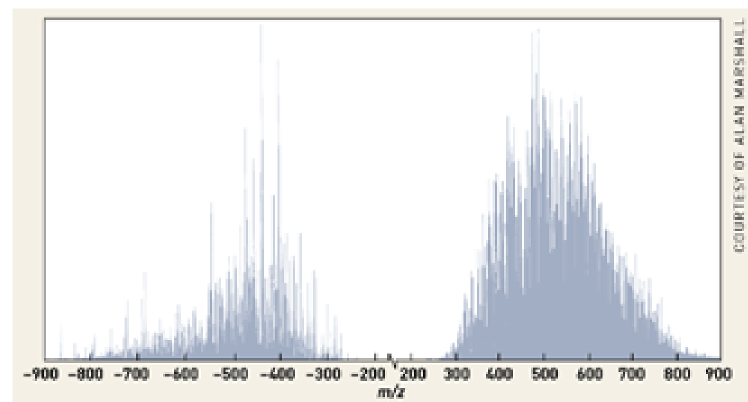
Why high magnetic fields?

- Induce new states of matter
- Destroy states of matter
- → Probe materials
  Fermi surfaces, NMR, MRI, spin levels with ESR, ion cyclotron resonance, etc.



Fermi surface of k-$(ET)_2Cu(NCS)_2$





Molecular mass

Mass spectrometry yields “Fingerprint” of South American crude oil

And most importantly….

MAGNETS CAN MANIPULATE MORALITY

Magnetic fields targeting the moral center of the brain could scramble our sense of right and wrong.

By Eric Bland
Mon Mar 29, 2010 03:01 PM ET
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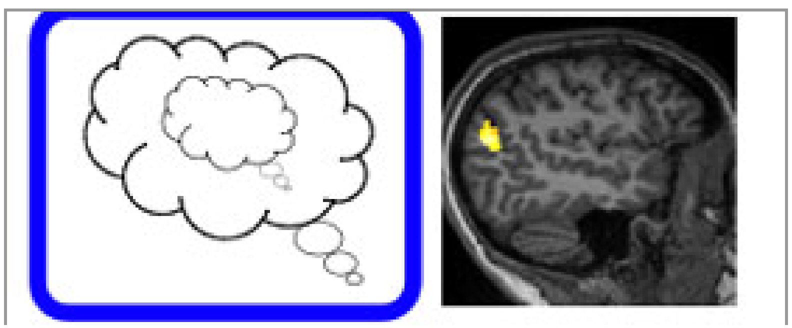


THE GIST:

- **Strong magnetic fields could affect moral judgment.**
- **Targeted magnetic fields can make people more inclined to judge outcomes, not intentions.**
- **The findings could have implications for neuroscience, as well as the legal system.**

Dr. Liane Young, MIT, PNAS 2010

Current state of the art:

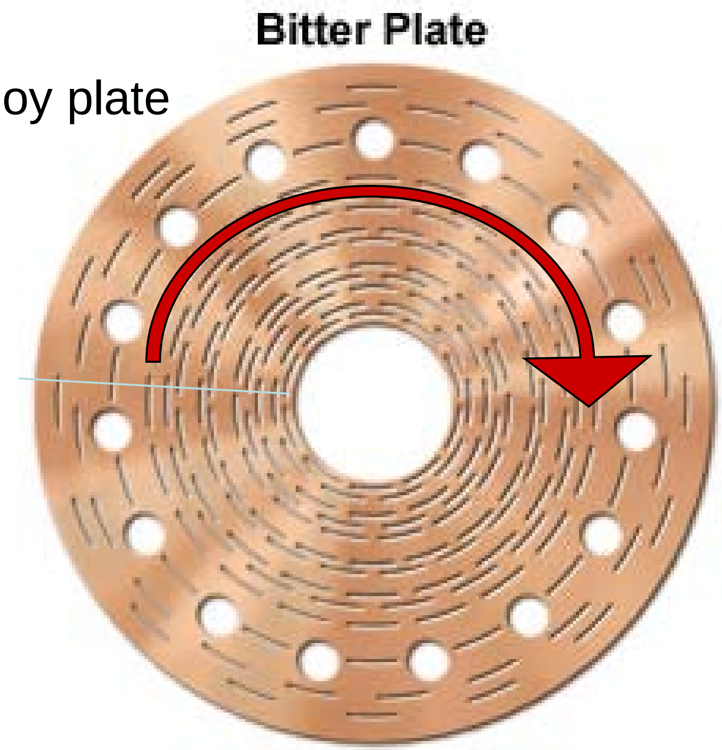
Superconducting magnets to 25 T

Complete 5 Tesl
with layer-wound

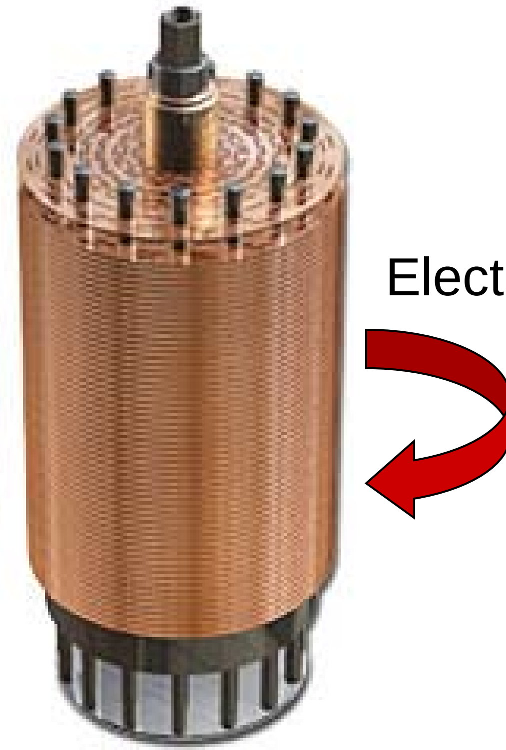
20 T Nb_3Sn magnet
+ 5 T high-T_c Bi2212 insert

Resistive “Florida-Bitter” Magnets up to 35 T

Copper-alloy plate



Electric current



Francis Bitter

Holes allow cooling water to flow
Staggered arrangement (“Florida” design) improves strength, max magnetic field

The worlds largest DC magnetic field: 33 T resistive + 12 T superconducting

Standing on the hybrid platform

Liquid helium plume

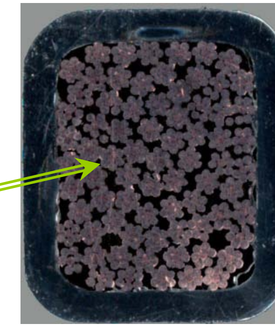
Field center 1.3 m

32 mm bore

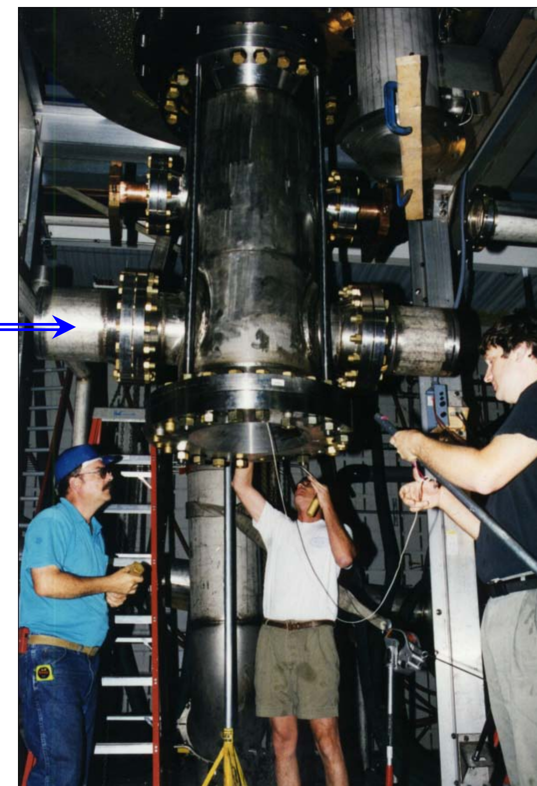
Cryostat designed to handle a fault load of 6 MN ≈ 27 times the thrust of a Boeing 747

45 Tesla

Cu/Nb_3Sn composite strands around Cu cores inside 10mm x 12mm steel conduit



8,000 liters of cooling water per second

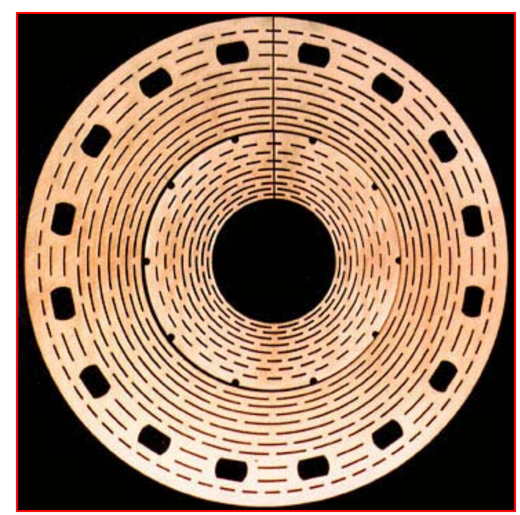


Resistive “Florida-Bitter” Magnets

30 foot tall cooling tower
(runs two 33 T magnets at a time)

Power bill ~ 6 million $ per year

NHMFL in Tallahassee, Florida



Here at LANL: Pulsed Magnets

Duration: ms to tens of ms instead of hours

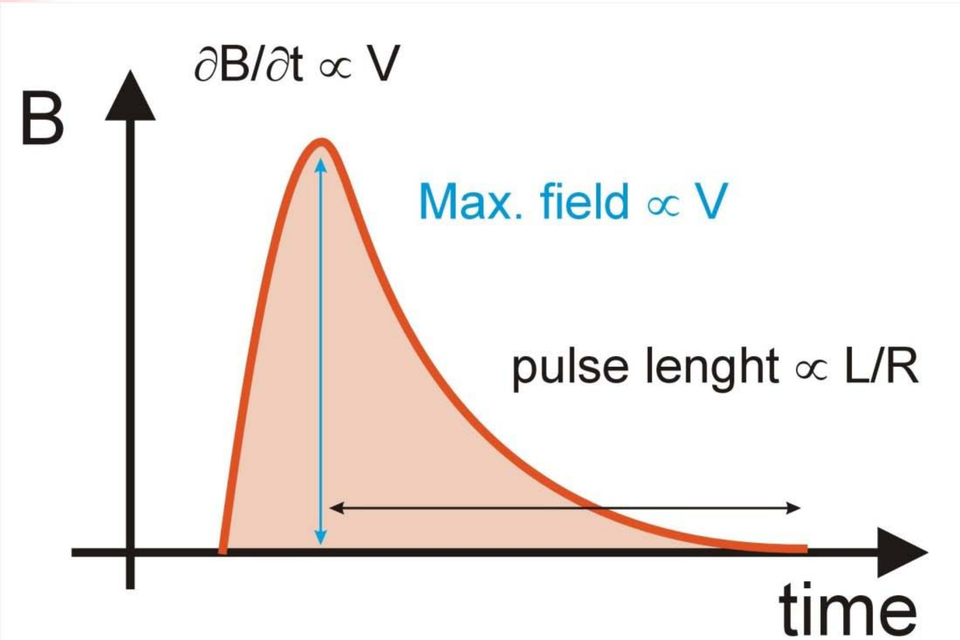
Higher peak magnetic fields

Much less power

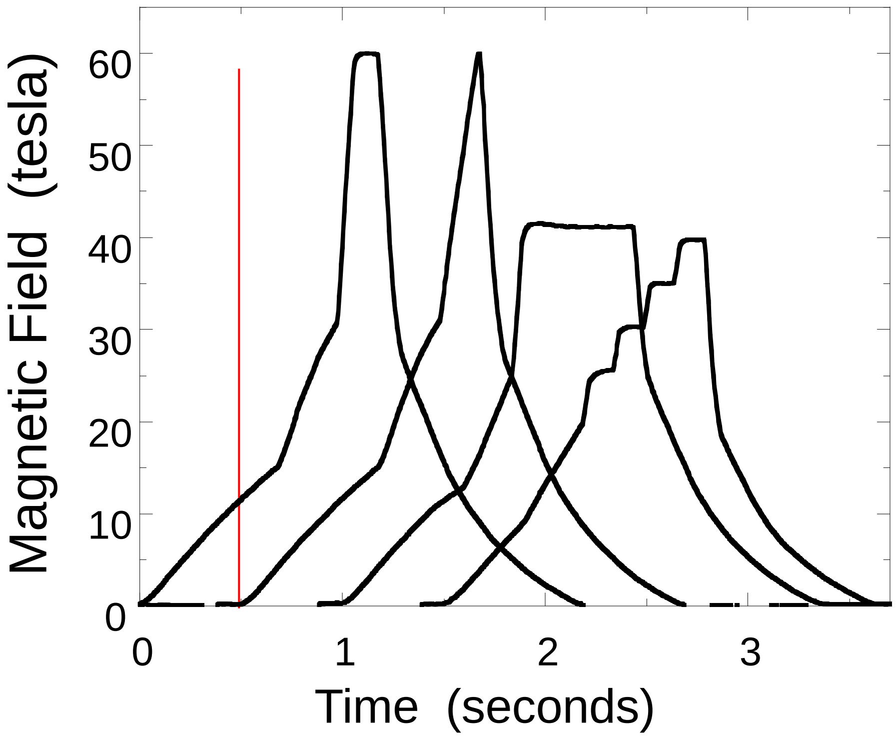
Cooling achieved by immersion in liquid nitrogen

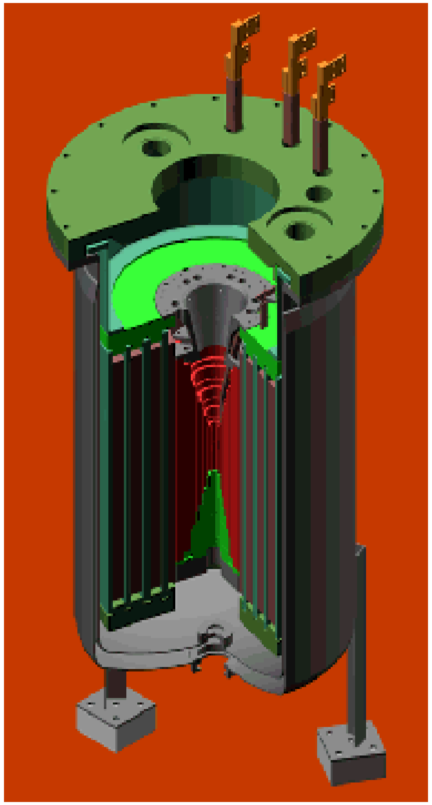
CuNb, CuAg, etc.

0.6 MJ of energy



60 T long pulse (world record)

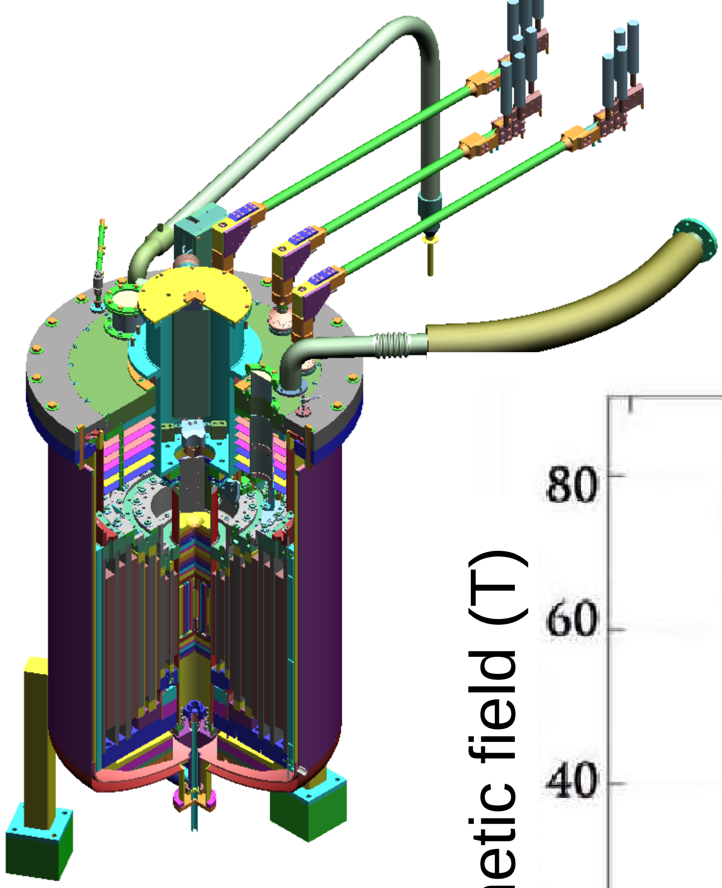


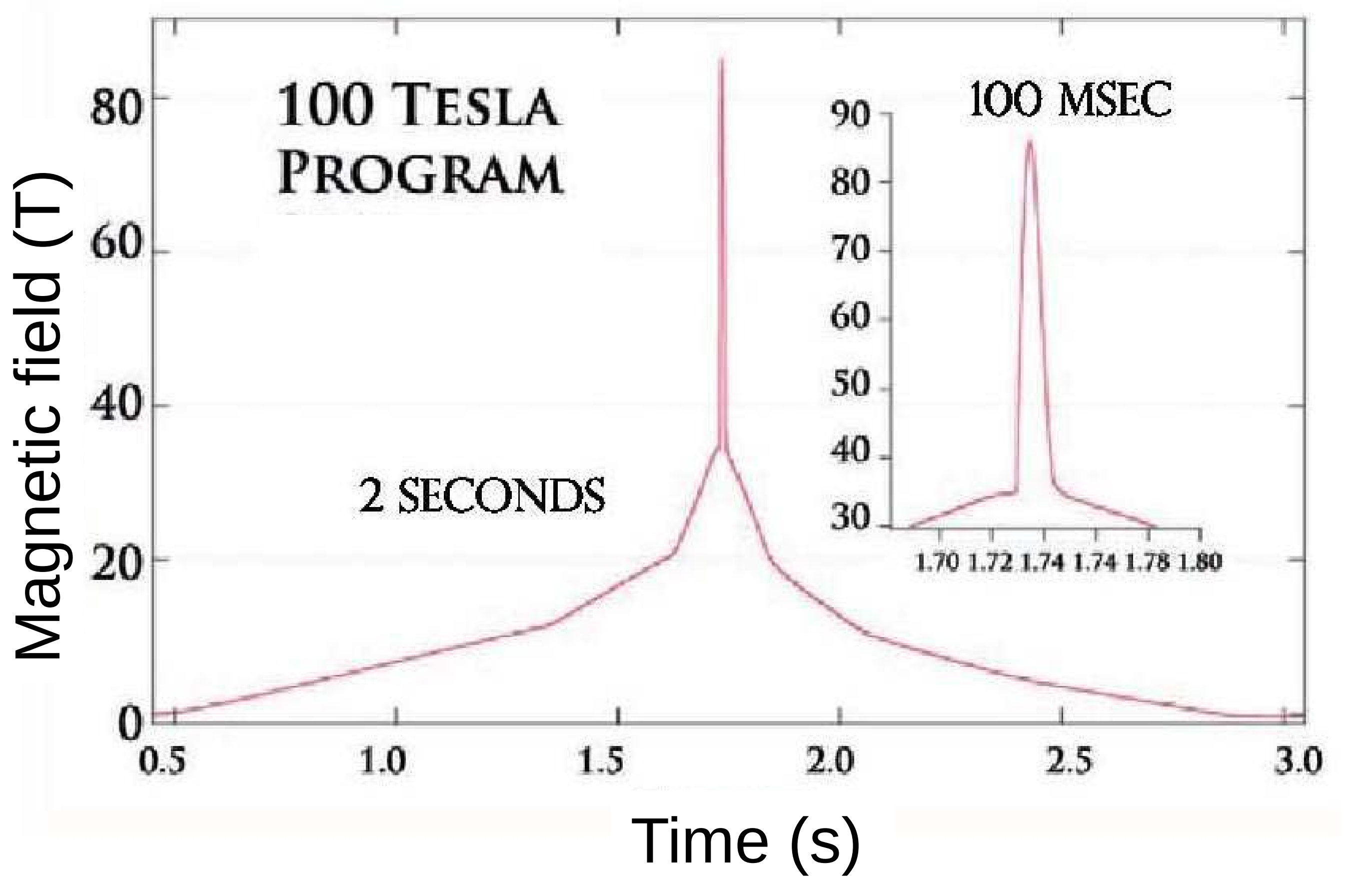


2 second pulse duration
1/10 second at peak field

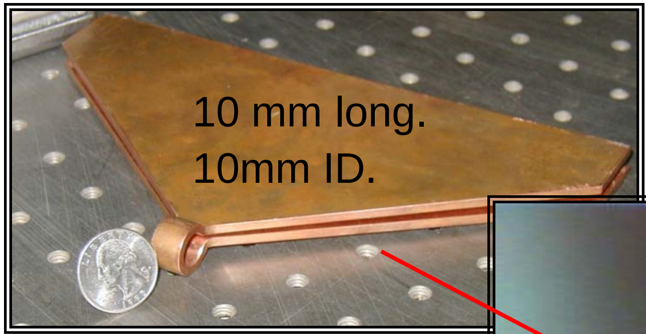
100 T multishot

(97.4 T so far)

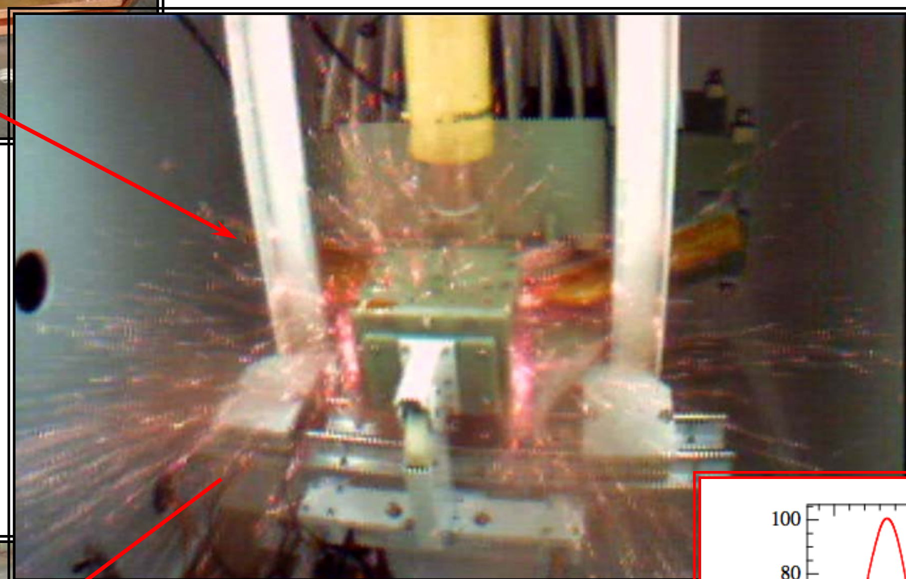




At 240 T, destroy the magnet every time, but not the sample



Capacitor bank pulses a short (μs) mega-amp current pulse to achieve ultra high magnetic fields.



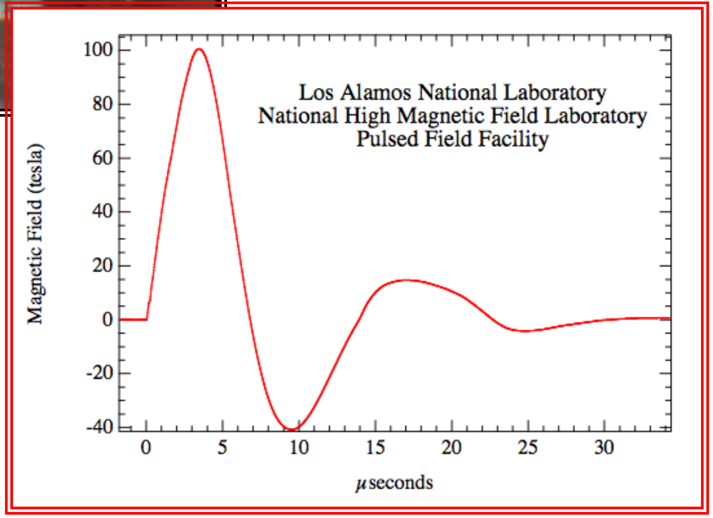
Low inductance capacitor bank.
L = 18 nH, C = 144 μF,
V = 60 kV, E = 259kJ.

Single turn magnet coil,
L = 7 nH.

Peak current 4 MA.

1st megagauss shot
(February 8th 2005)

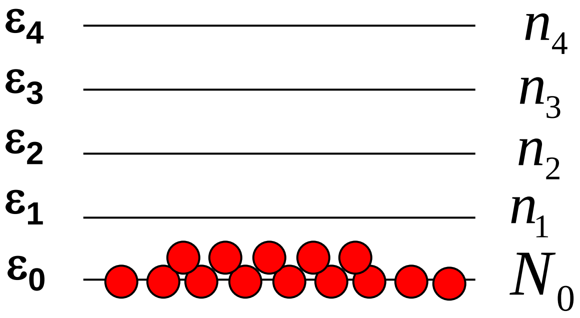
$\Rightarrow$ 2 μs rise time,
$\Rightarrow$ dB/dt ≈ 10^8 Ts^{-1}
3 orders of magnitude faster than standard short pulse magnets at the NHMFL.



Bose-Einstein condensation

Ingredients:

1. Bosons
2. Number conservation of the bosons
3. Coherent wave function

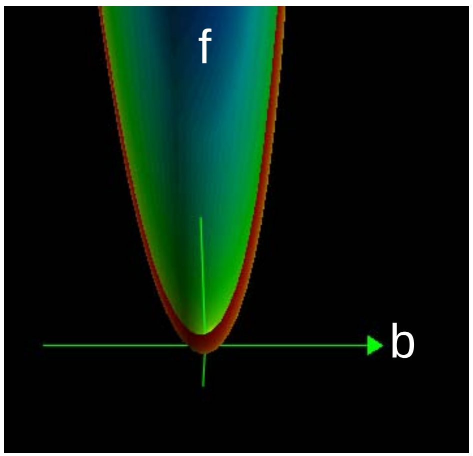


Lower temperature, bosons forced to condense into the ground state

Mapping BEC onto spins

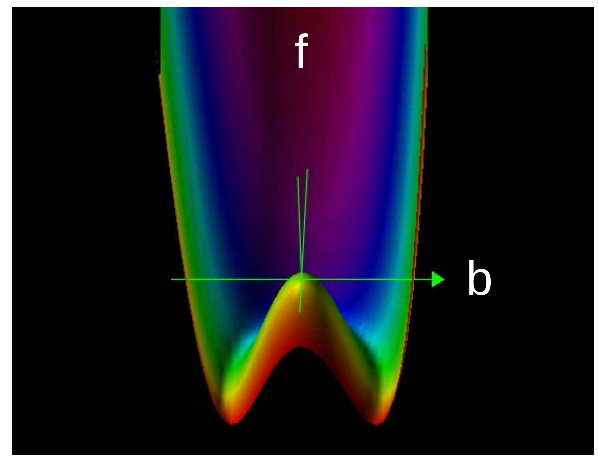
Number conservation is synonymous with U(1) symmetry

U(1) symmetry = axial symmetry



U(1) symmetry

Phase transition
(T,H,P, …)



Spontaneous symmetry breaking picks an angle ϕ

BEC transition = spontaneous U(1) symmetry breaking

Order parameter has magnitude AND phase **$be^{i\phi}$**

(Universality class ν = 2/3, z = 2)

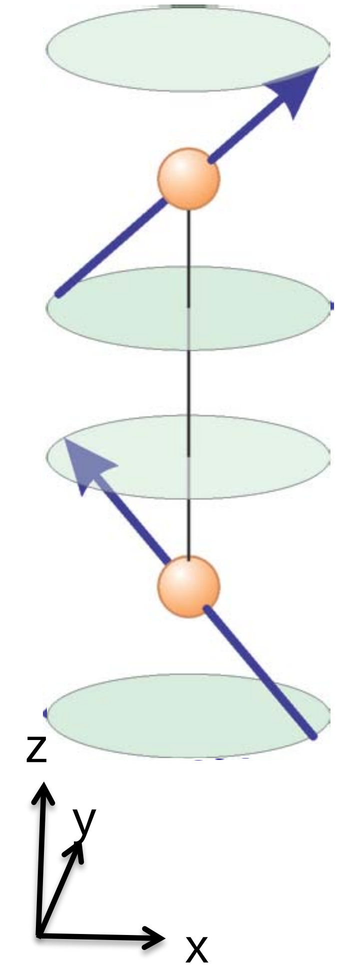
Mapping BEC onto spins

U(1) symmetry = XY magnetism

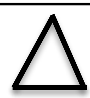
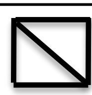
Transition into BEC =
formation of long-range XY AFM, with spontaneous choice of axial angle

Transition can be tuned by temperature or ***magnetic field***

Experimentally there is never a perfect U(1) symmetry (even in the atomic BECs). It's a matter of being in the right limit that the theroy applies.

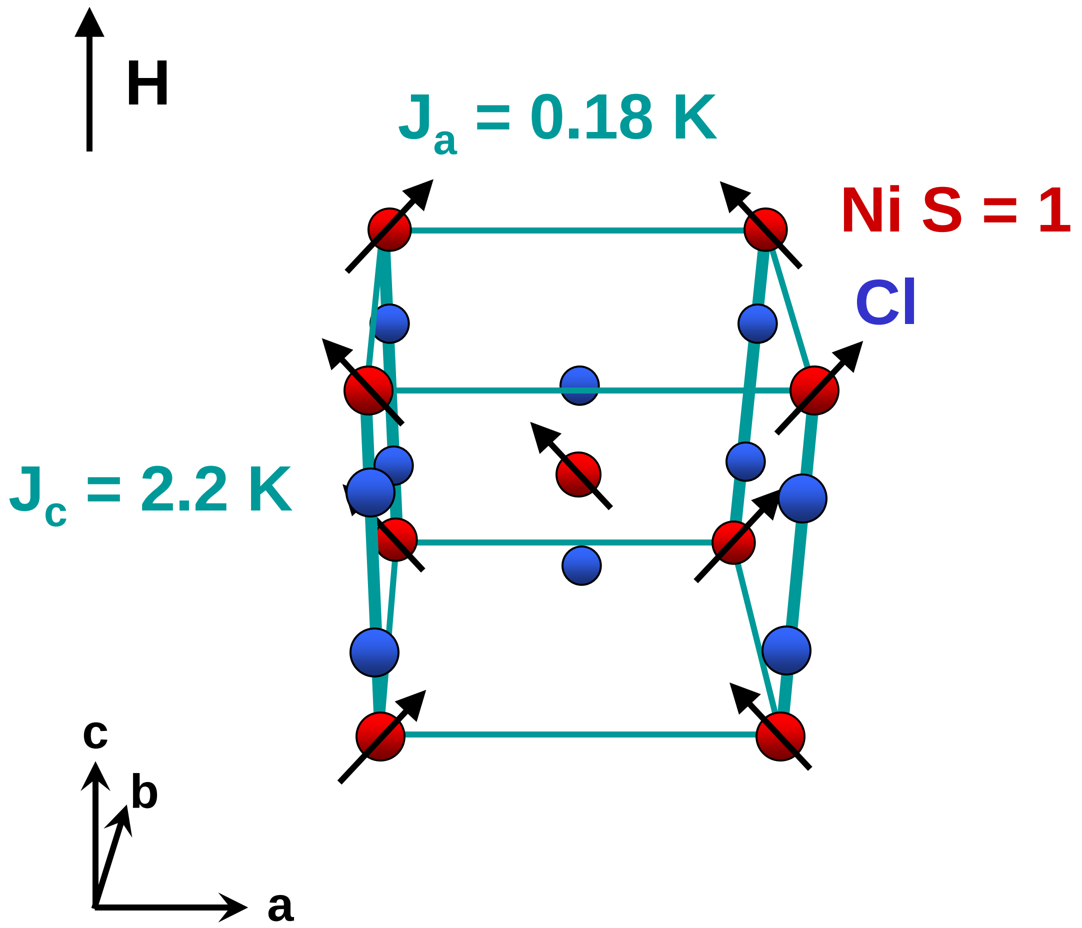


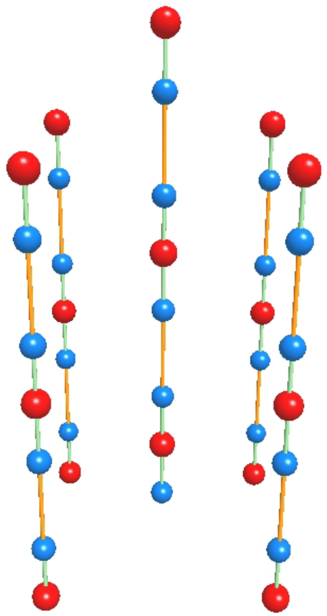
Partial list of BEC quantum magnets

Compound	**Spins**	**Max T_c(K)**	**H_{c1}, H_{c2}(T)**	**Spin gap**	**Crystal symmetry**
$BaCuSi_2O_6$	↓↑ S = 1/2	3.8 K	24 T, 49 T	5 meV	tetragonal
$TlCuCl_3$ *anisotropy found	↓↑ S = 1/2	> 8 K	5 T, ~100 T	6 meV	monoclinic
$KCuCl_3$	↓↑ S = 1/2	>5.5 K	23 T, 55 T	4 meV	monoclinic
$Ba_3Cr_2O_8$ *anisotropy found	↓↑ S = 1/2	2.7 K	13 T, 24 T	1.6 ,2.2 meV	rhombohedral
$Pb_{.2}V_3O_9$	↓↑ S = 1/2	4 K	4 T, ~40 T	1 meV	triclinic
$(CH_3)2(CHNH_3CuCl_3)$ [IPA-$CuCl_3$]	↑↑ S = 1/2	10 K	10 T,?	1 meV	triclinic
Cs_2CuCl_4	 S = 1/2	0.32 K	N/A, 9 T	0	orthorhombic
$(CuCl)LaNb_2O_7$	 S = 1/2	>3.3 K	9 T, ?	2 meV	orthorhombic
$NiCl_2$-$4SC(NH_2)_2$ [DTN]	↑ S = 1	1.2 K	2 T, 13 T	1 meV	tetragonal
$Ba_3Mn_2O_8$	↓↑ S = 1	0.87; 0.65 K	9 T, 26 T; 32 T, 48 T	2 meV	rhombohedral
F_2PNNNO	↓↑ S = 1	1.5 K, -	10 T, 15 T; 26 T,29 T	1 meV	orthorhombic

$NiCl_2$-$4SC(NH_2)_2$ (DTN)

(thiourea molecules omitted)

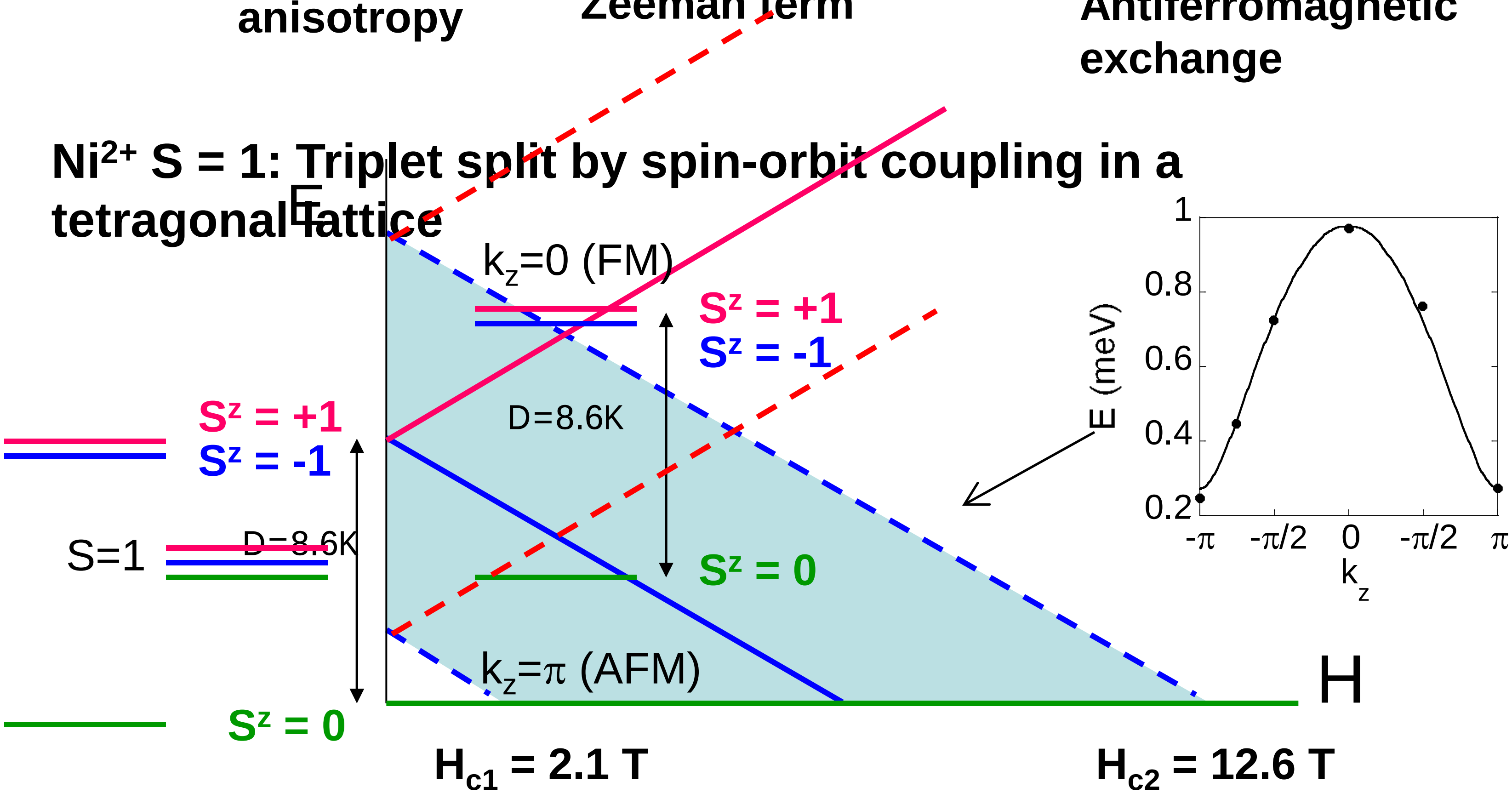


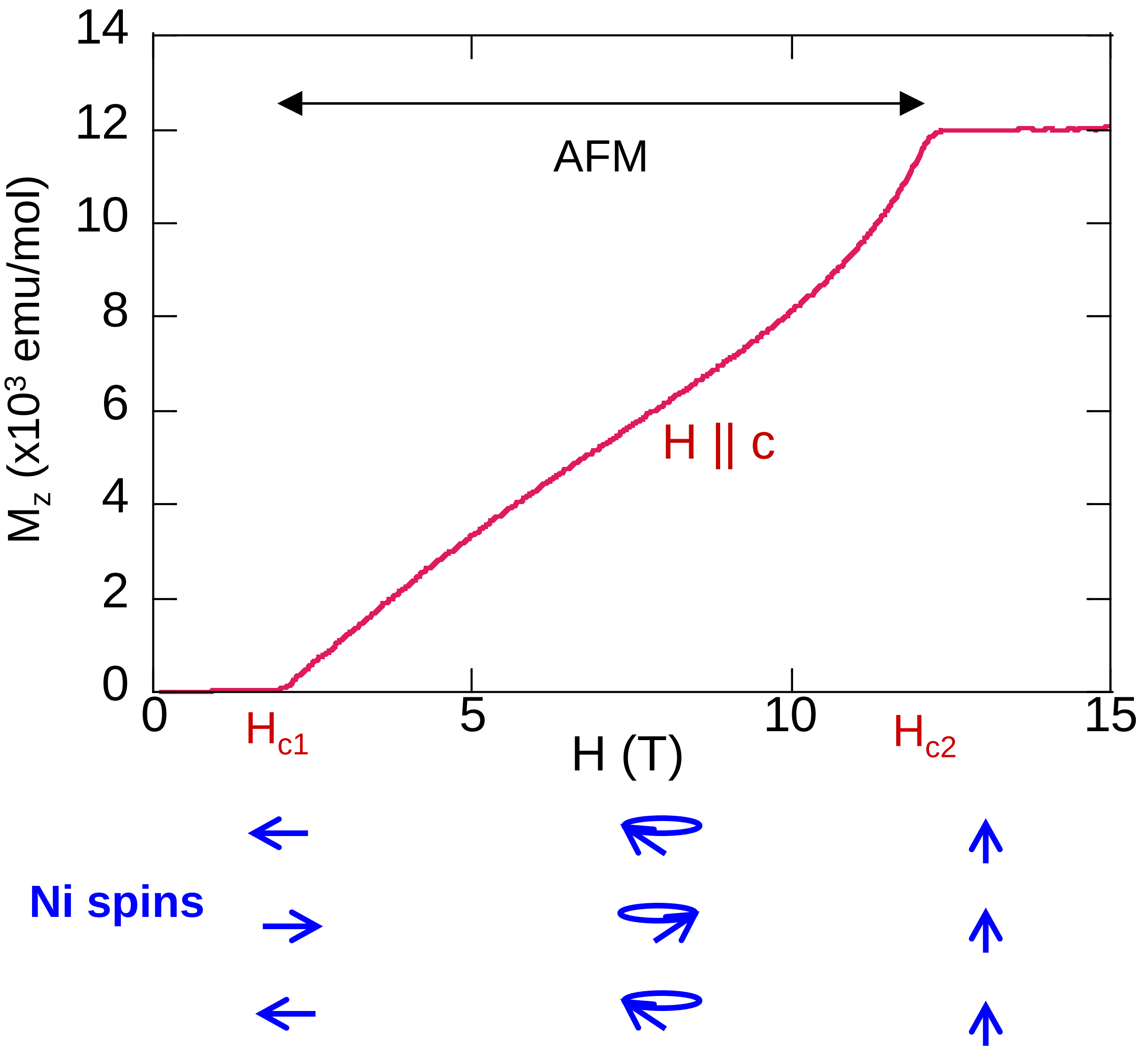


$$H = D\sum_j \left(S_j^z\right)^2 - g\mu_B H_z \sum_j S_j^z + \sum_{\nu<ij>} J_\nu \vec{S}_i \cdot \vec{S}_j$$

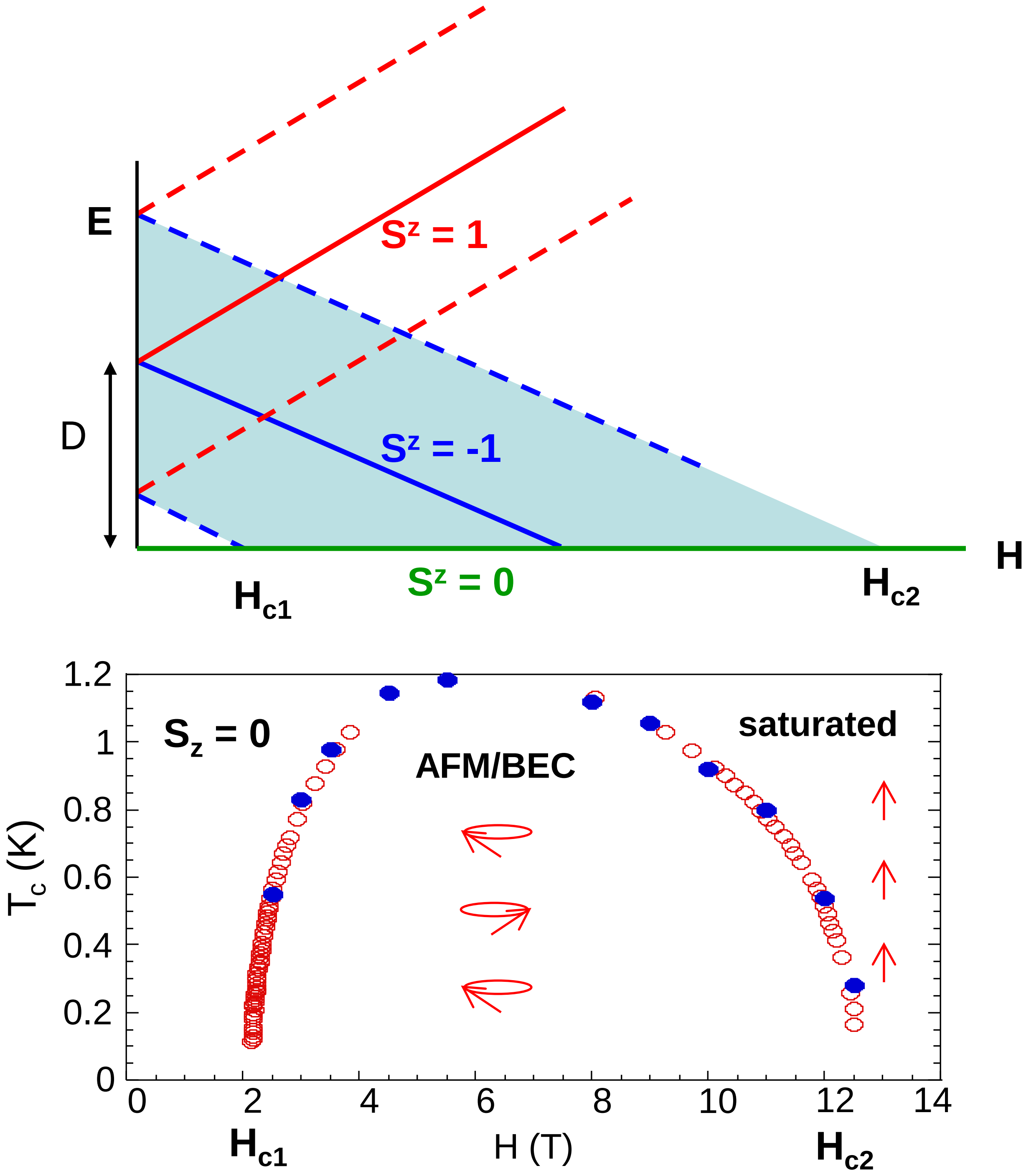
Uniaxial anisotropy | **Zeeman term** | **Antiferromagnetic exchange**

Ni^{2+} S = 1: Triplet split by spin-orbit coupling in a tetragonal lattice





A. Paduan-Filho et al, Phys. Rev. B 69, 020405(R) (2004)


E
S^z = 1
D
S^z = -1
H
S^z = 0
H_c1
H_c2
1.2
1
0.8
0.6
0.4
0.2
0
S_z = 0
saturated
AFM/BEC
T_c (K)
0
2
4
6
8
10
12
14
H_c1
H (T)
H_c2

Spin language Hamiltonian

$$\mathrm{H} = D\sum_i \left(S_i^z\right)^2 - g\mu_B H \sum_i S_i^z + \sum_{<i,j>\nu} J_\nu \vec{S}_i \cdot \vec{S}_j$$

Spin-orbit coupling | **Magnetic field/Zeeman term** | **AFM exchange**

$S^+ \rightarrow b^+$ (Creation operator for $S_z = 1$ or a boson)
Subject to semi-hard-core constraint of 2 bosons/site

Boson language Hamiltonian (neglecting $S_z = -1$ term)

$$\mathrm{H} = \sum_{<i,j>\nu} J_\nu \left(b_i^+ b_i b_j^+ b_j + b_i^+ b_j + b_j^+ b_i\right) + h_{eff}(D)\sum_i b_i^+ b_i$$

Repulsion (2nd order in N) | **hopping** | **number operator**

Constraint: One boson per site

2. Boson number conservation: Axial symmetry

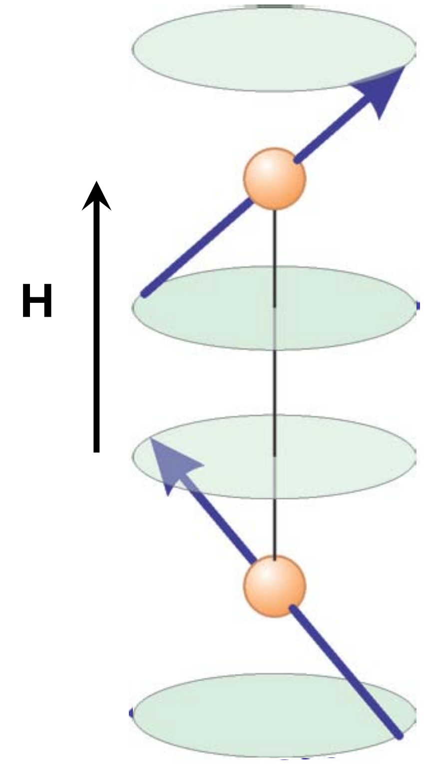
Consider boson creation operator $\mathbf{b}^\dagger$

$\mathbf{b}^\dagger \rightarrow \mathbf{b}^\dagger\, \mathbf{e}^{\mathbf{i}\phi}$ under axial rotation

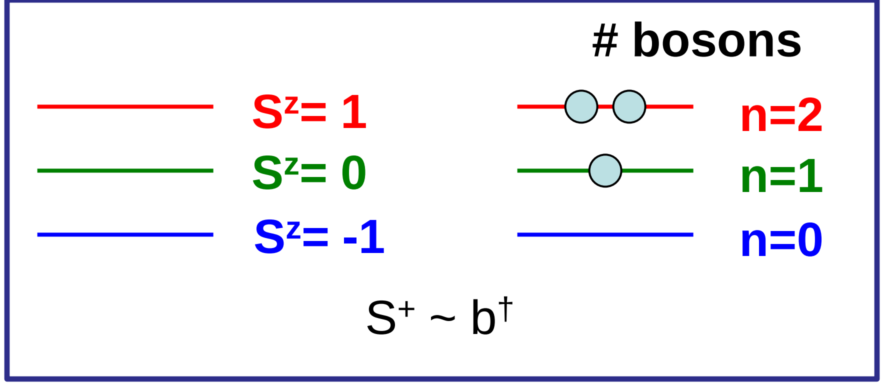
Now consider number operator $\mathbf{N}= \mathbf{b}^\dagger\, \mathbf{b}$

$\mathbf{b}^\dagger\, \mathbf{b} \rightarrow (\mathbf{b}^\dagger\, \mathbf{e}^{\mathbf{i}\phi})\, (\mathbf{b}\mathbf{e}^{-\mathbf{i}\phi}) = \mathbf{b}^\dagger\, \mathbf{b} = \mathbf{N}$

Number operator is conserved under axial rotation



$$\mathrm{H} = \sum_{<i,j>\nu} J_\nu \left(b_i^+ b_i b_j^+ b_j + b_i^+ b_j + b_j^+ b_i \right) + h_{eff}(D) \sum_i b_i^+ b_i$$



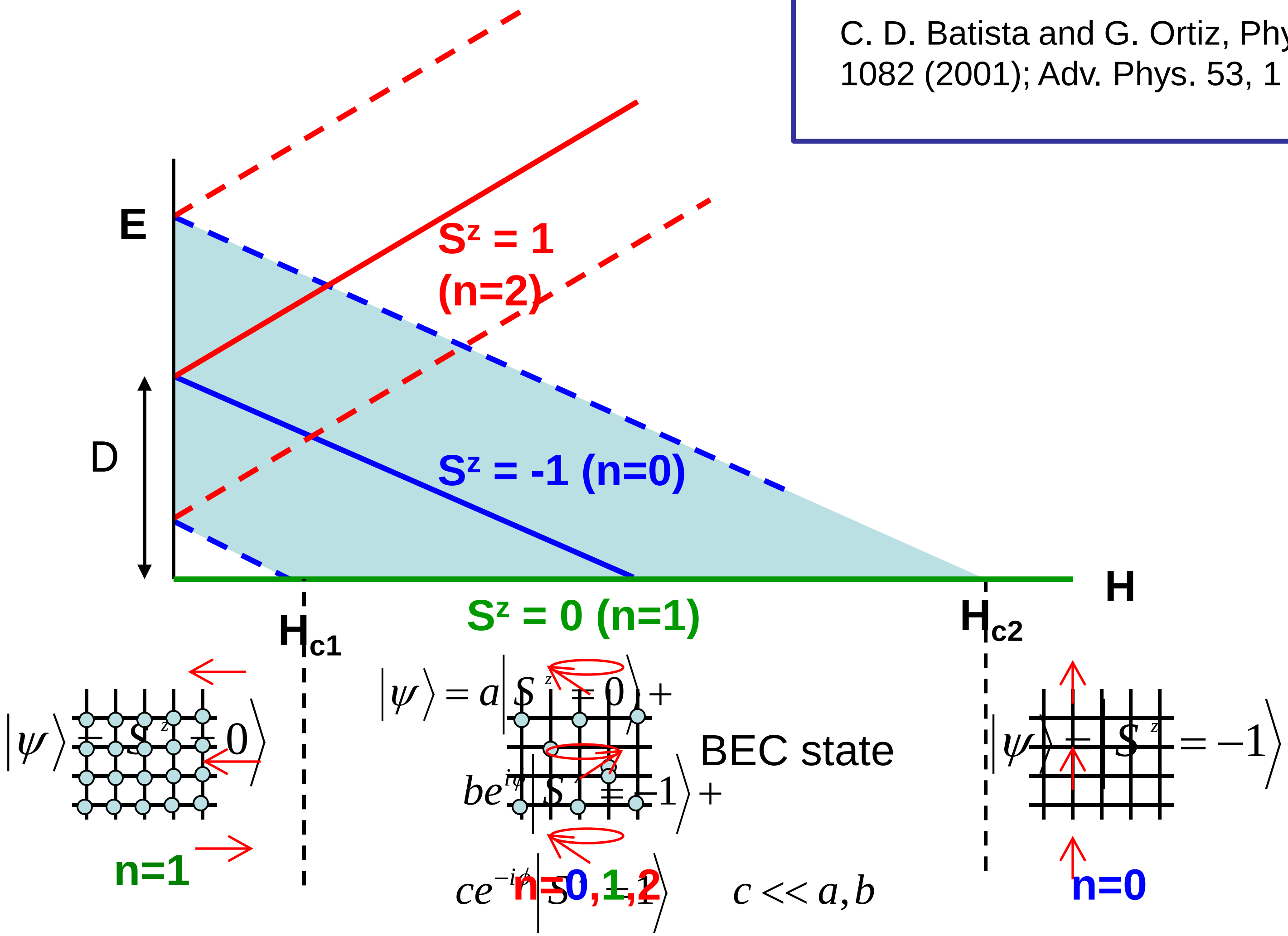
Bosonic Mott-Hubbard model

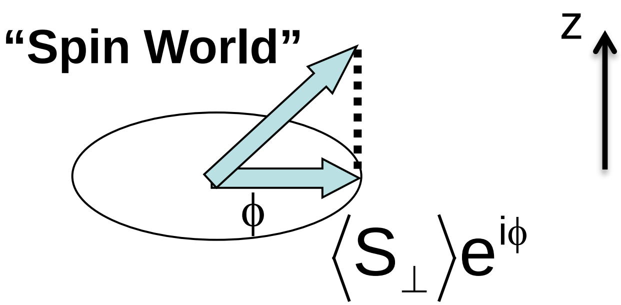
bosons ~ M_{sat}-M

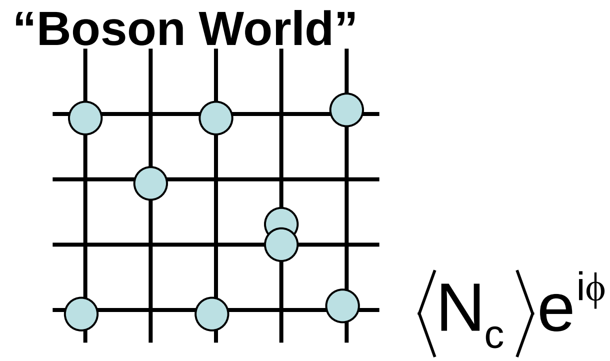
condensed bosons ~ staggered M_{xy}

H ~ μ (chemical potential)

C. D. Batista and G. Ortiz, Phys. Rev. Lett. 86, 1082 (2001); Adv. Phys. 53, 1 (2004).

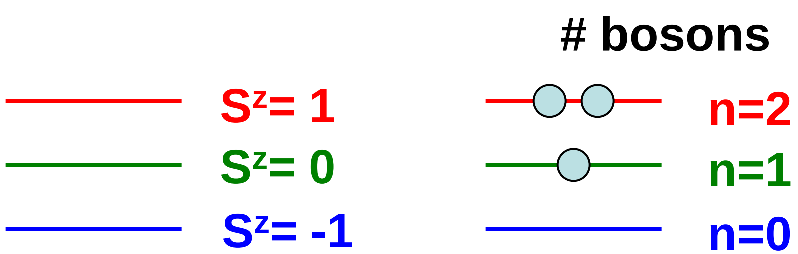






Order parameter:
Staggered magnetization in XY plane
(Magnitude and phase)

Order parameter:
Number of condensed
bosons

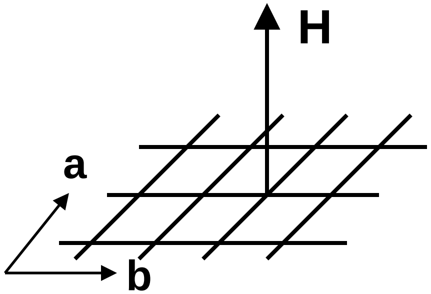


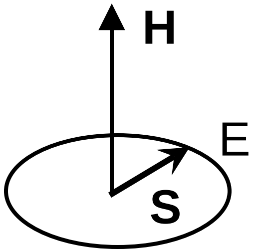
# of bosons = S_z
condensed bosons = S_{xy}

Limitations of the BEC description of magnets

Conservation of boson number is violated on short time scales because the magnetization fluctuates

-> BEC description valid only in equilibrium





Effects that break uniaxial symmetry,

allow the magnetization to fluctuate

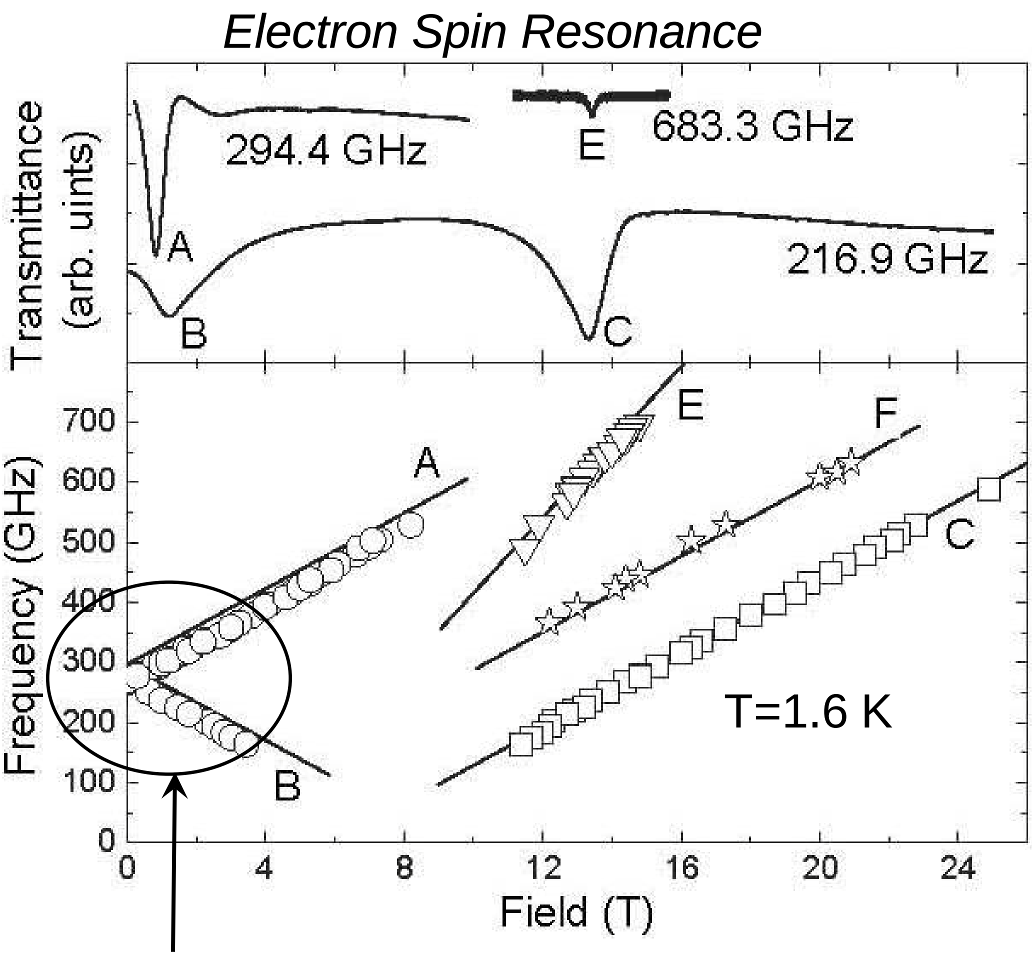
- **Diagonal spin-orbit coupling (e.g. spins see the lattice structure)**
- **Off-diagonal spin-orbit couplings (e.g. Dzyaloshinskii-Moriya)**
- **Non-tetragonal structural distortions**
- **Dipole-dipole interactions**

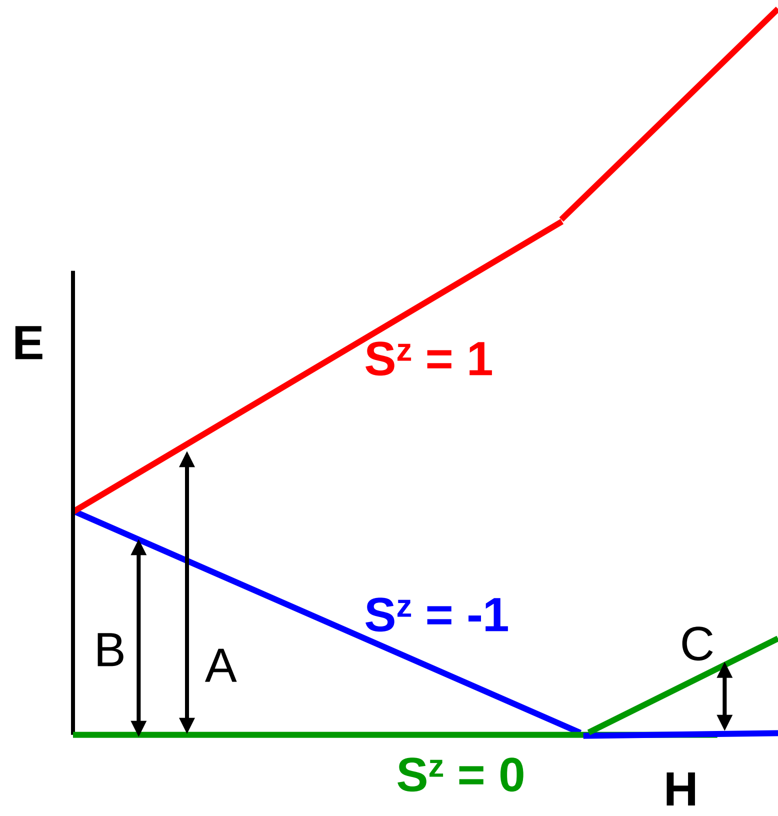
Our goal: Find compounds in which anisotropy (<μK) is much smaller than T_{BEC} (K)

Advantages of BEC description of certain magnets

- Magnets provide an experimental BEC in the thermodynamic limit
- Formalism and results of the theory of weakly interacting bosons can be transplanted wholesale to help understand quantum magnetism
- Transforming the Hamiltonian to boson language can greatly simplify the math
- Understanding quantum magnetism of localized spins is the first step before adding itinerant electrons
- Quantum magnetism underlies high-T_c cuprates, multiferroics, many other subjects

Experimental evidence of BEC and axial symmetry

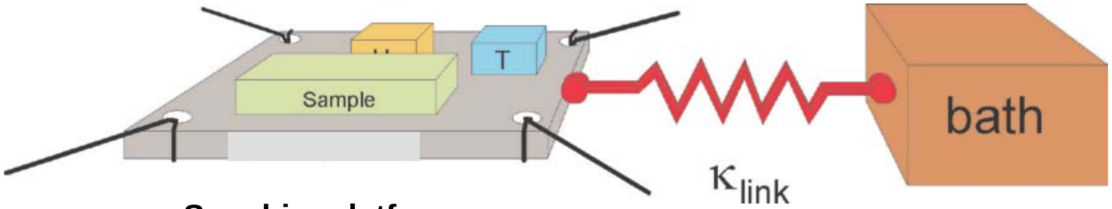




Straight lines and intersection of A and B confirms axial symmetry/BEC

S. Zvyagin, et al, *Phys. Rev. Lett.* 98, 047205 (2007).

Measuring Specific Heat
Of $NiCl_2$-$4SC(NH_2)_2$



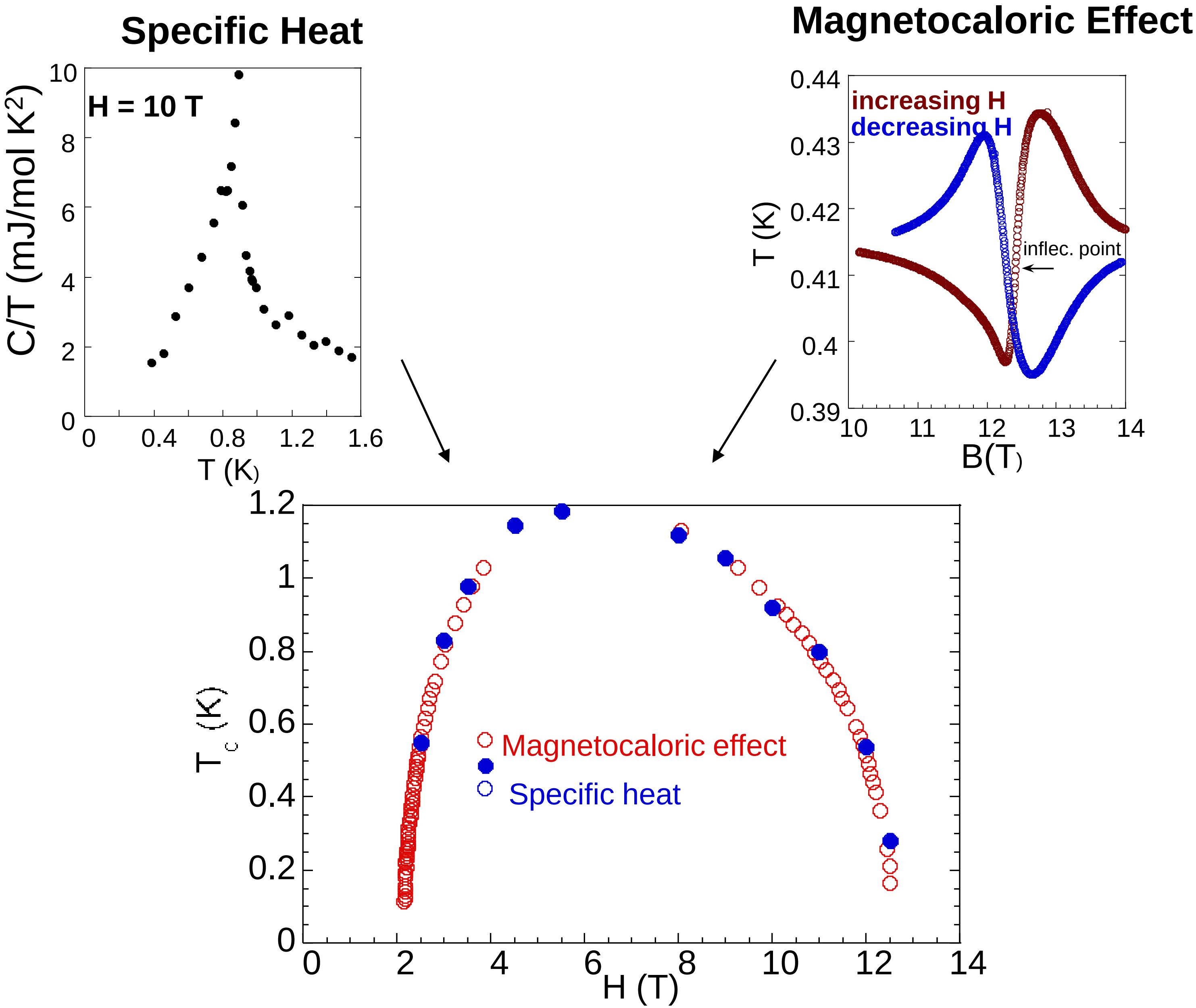
Sapphire platform

$C = Q/\Delta T$

Quasi-Adiabatic

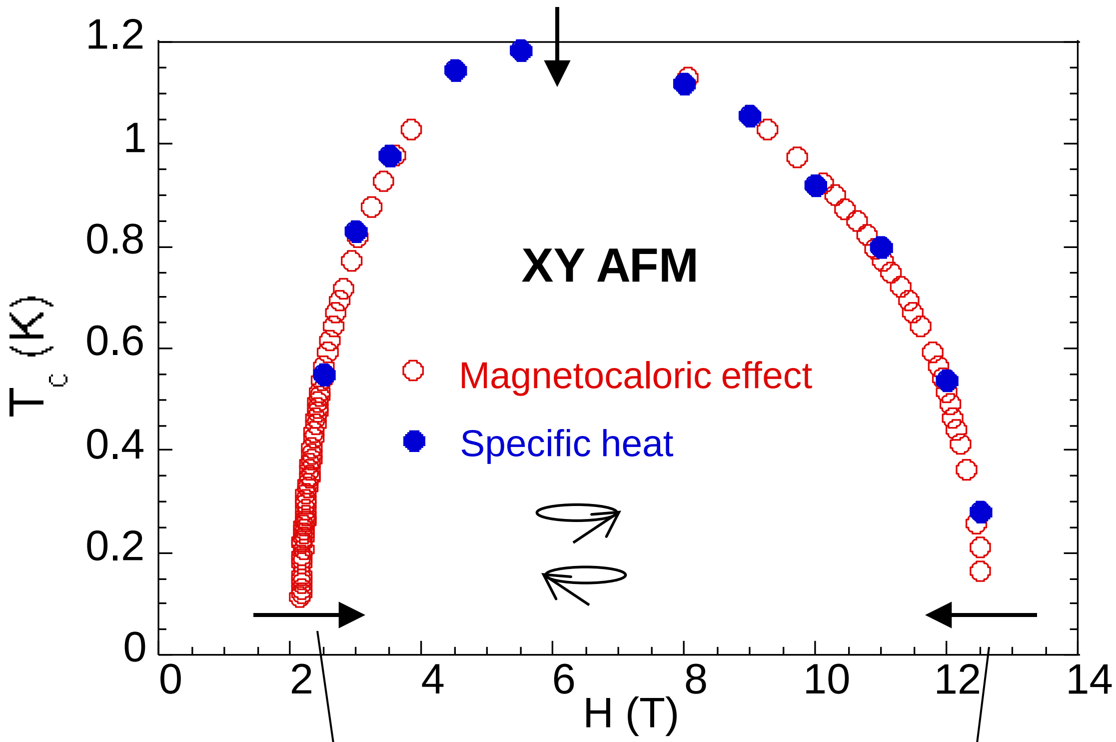
$C = \tau/\kappa$

Thermal Relaxation Time



V. S. Zapf et al, Phys. Rev. Lett., 96, 077204 (2006)

Thermal phase transition (XY AFM)
phase decoherence only (d=3)



Quantum Phase Transition (BEC)
amplitude and phase (d=3, z=2)

$M_z \propto T^{\alpha}$	3D BEC: α = 3/2
	3D Ising: α = 2
$H_c - H_{c1} \propto T^{\alpha}$	2D BEC: α = 1

$$M_z \propto T^{\alpha}$$

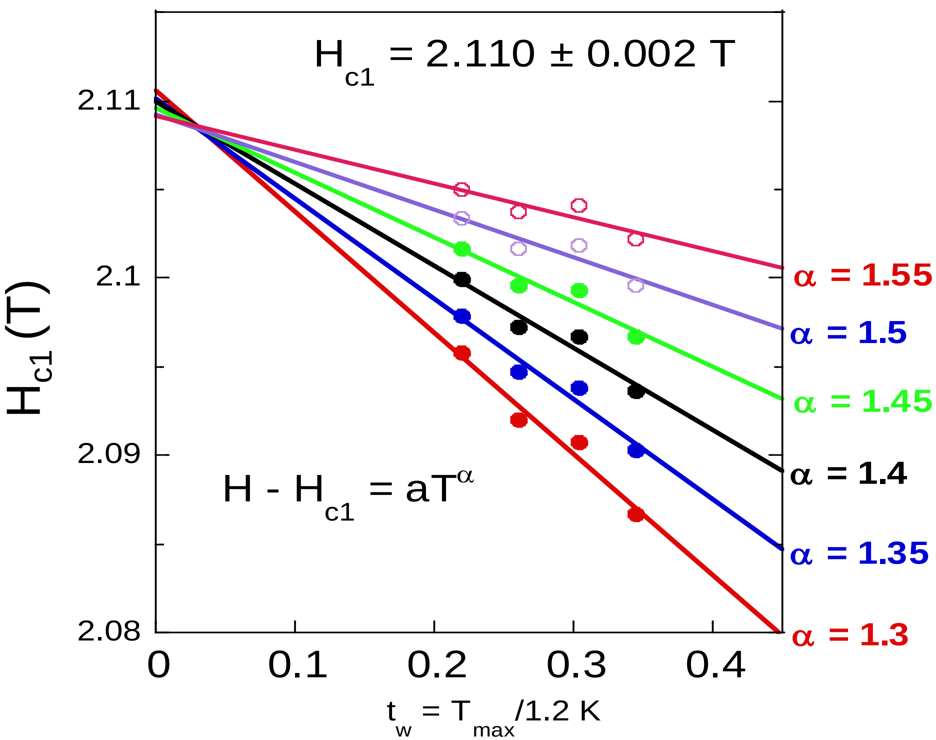
$$H_c - H_{c1} \propto T^{\alpha}$$

3D BEC: $\alpha = 3/2$

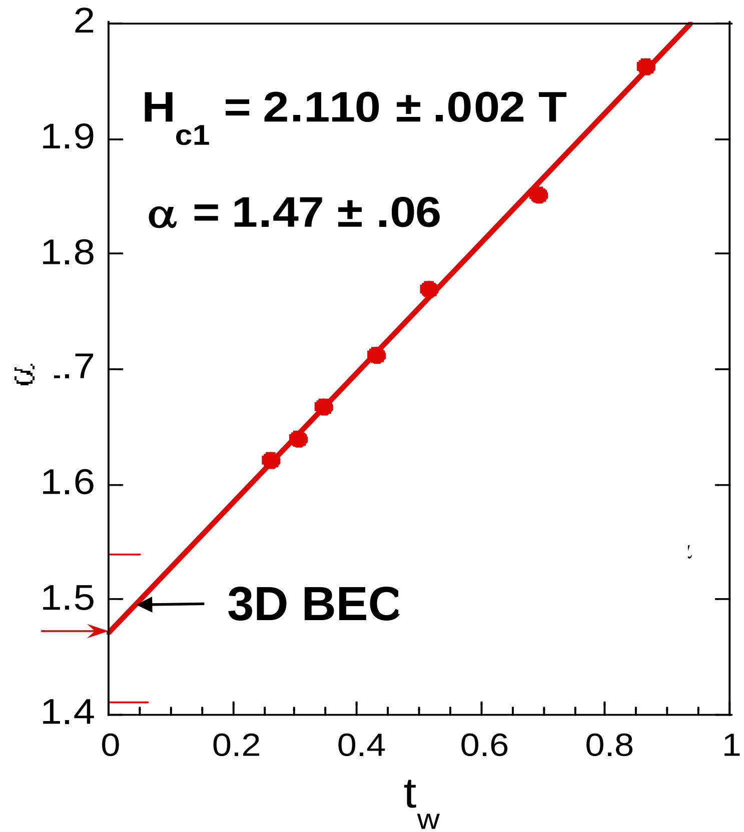
3D Ising: $\alpha = 2$

2D BEC: $\alpha = 1$

Windowing technique
Problem: exponent expected at T = 0



1. Fix α, fit to determine H_{c1}



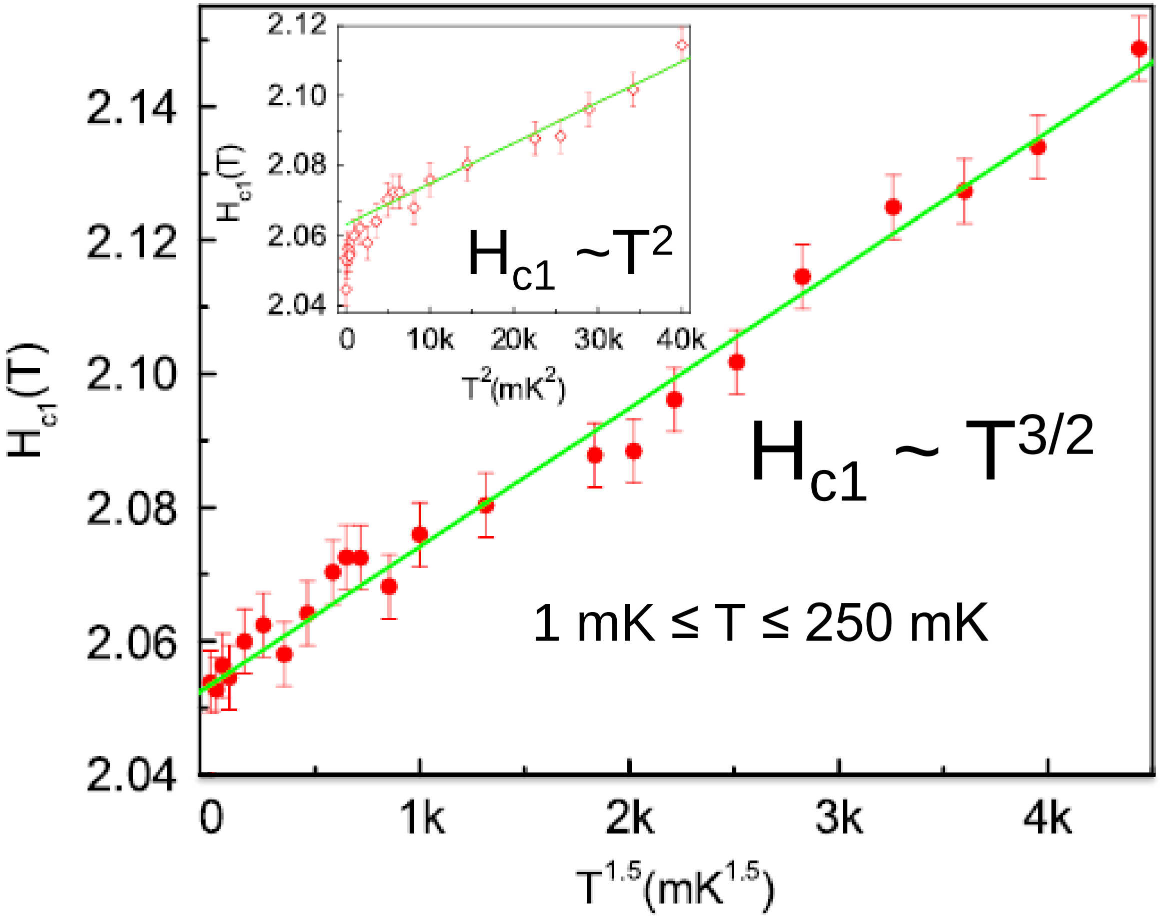
2. Using H_{c1}, determine α

S. Sebastian et al, **Phys. Rev. B 72, 100404(R) (2005) ($BaCuSi_2O_6$)**

V. S. Zapf, et al, **Phys. Rev. Lett., 96, 077204 (2006) (this compound)**

O. Nohadani et al, **Phys. Rev. B 69 220402(R) (2004) (QMC)**

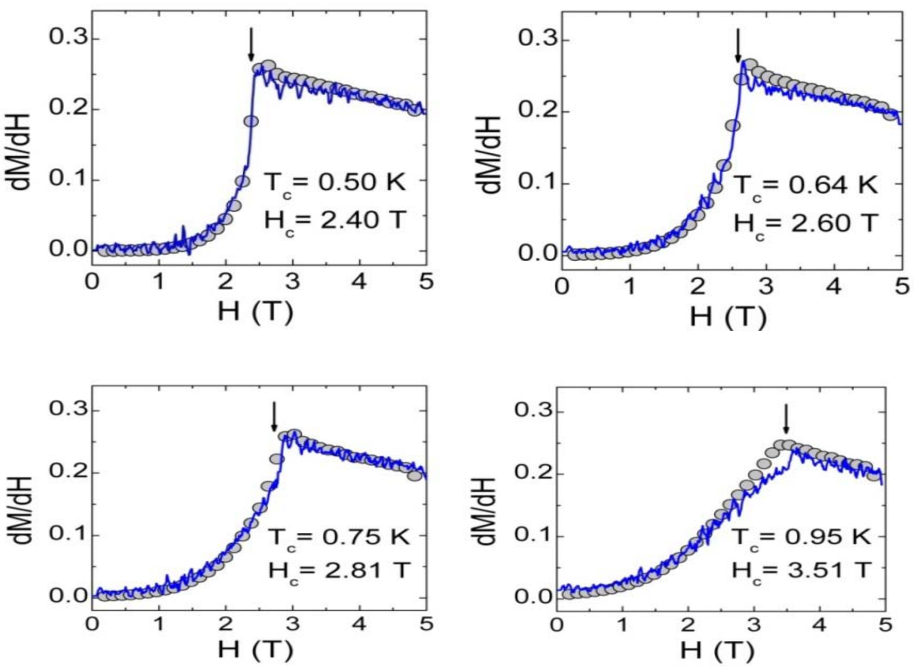
Direct measurement to 1 mK
Ac susceptibility

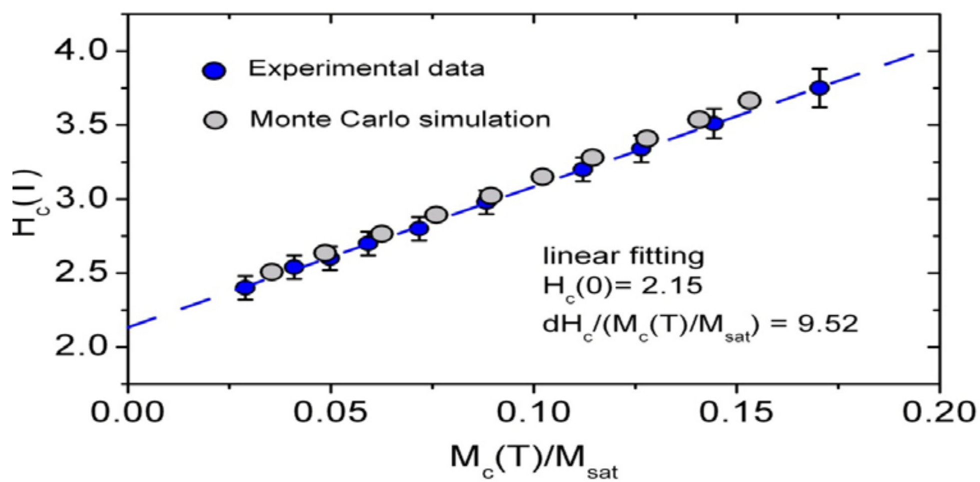


L. Yin, J.-S. Xia, V. S. Zapf, N. Sullivan, A. Paduan-Filho, *Phys. Rev. Lett*. **101**, 187205 (2008).

Data taken at high B/T Laboratory Gainesville, Florida

Power-law behavior of the magnetization





A. Paduan-Filho et al, *Phys. Rev. Lett.* **102**, 77204 (2009).

We hope we have now measured all the significant terms in this Hamiltonian

$$H = D\sum_j \left(S_j^z\right)^2 - g\mu_B H_z \sum_j S_j^z + \sum_{v<ij>} J_v \vec{S}_i \cdot \vec{S}_j$$

D = 8.9 K

J_c = 2.2 K

J_a = 0.18 K

g = 2.26

Inelastic Neutron diffraction: *D, J*

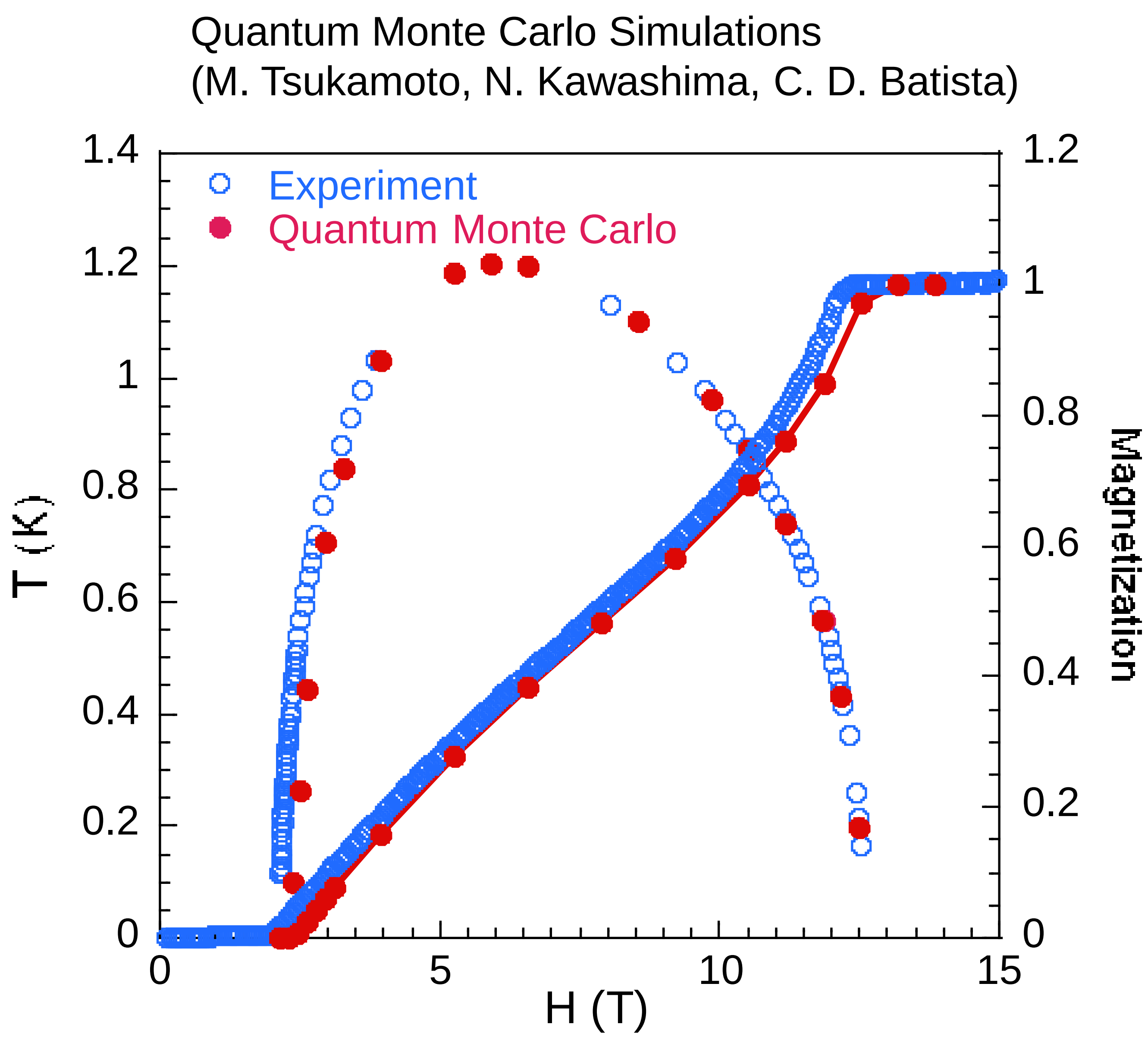
V. S. Zapf, et al, Phys. Rev. Lett., 96, 077204 (2006)

Electron spin resonance: *D, J*

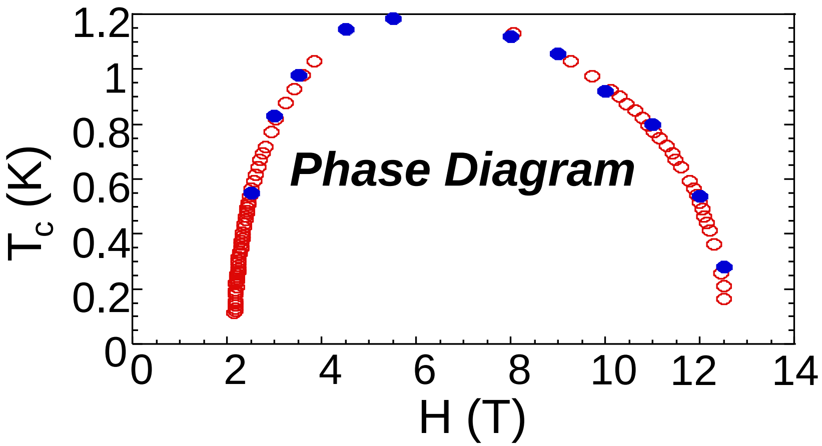
S. Zvyagin et al, Phys. Rev. Lett. 98, 047205 (2007)

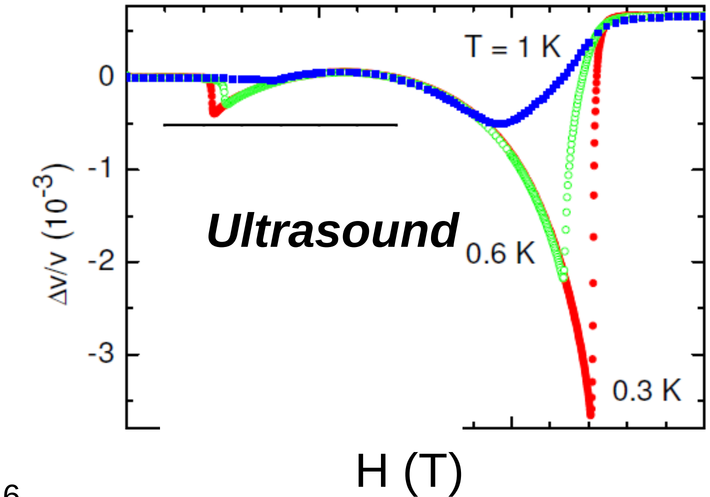
Magnetization, ESR: *g*

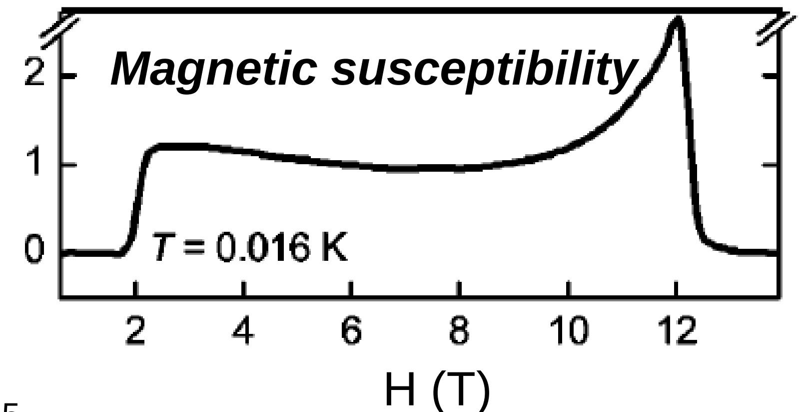
A. Paduan-Filho et al, Phys. Rev. B 69, 020405(R) (2004)

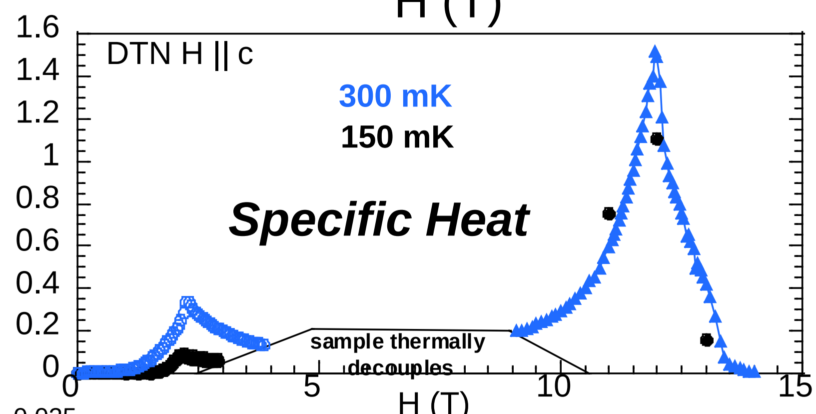

Quantum Monte Carlo Simulations
(M. Tsukamoto, N. Kawashima, C. D. Batista)
Experiment
Quantum Monte Carlo
T (K)
Magnetization
H (T)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
0
5
10
15
0
0.2
0.4
0.6
0.8
1
1.2

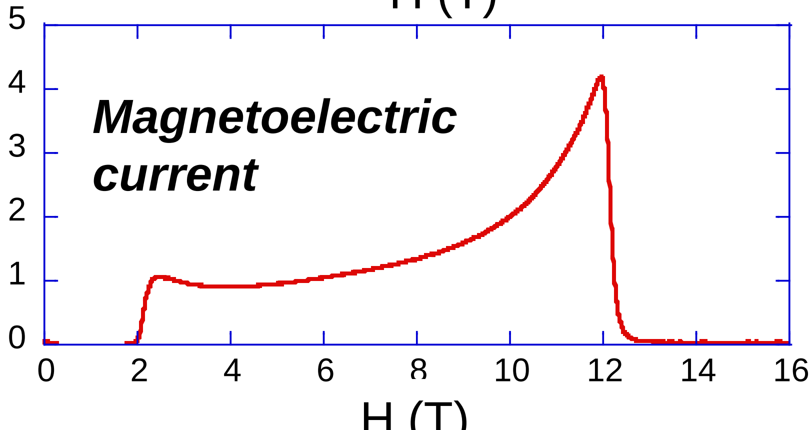
Asymmetry in many properties of DTN (and other BEC compounds)

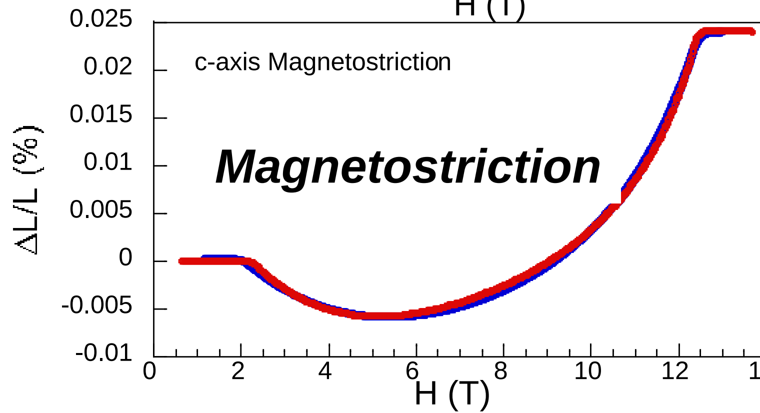




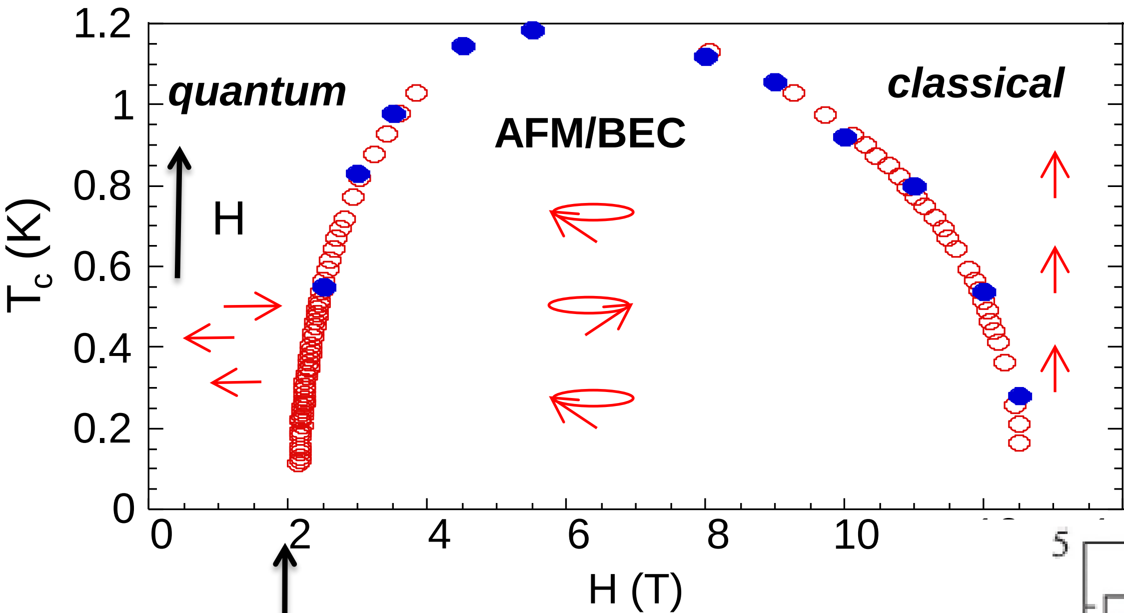








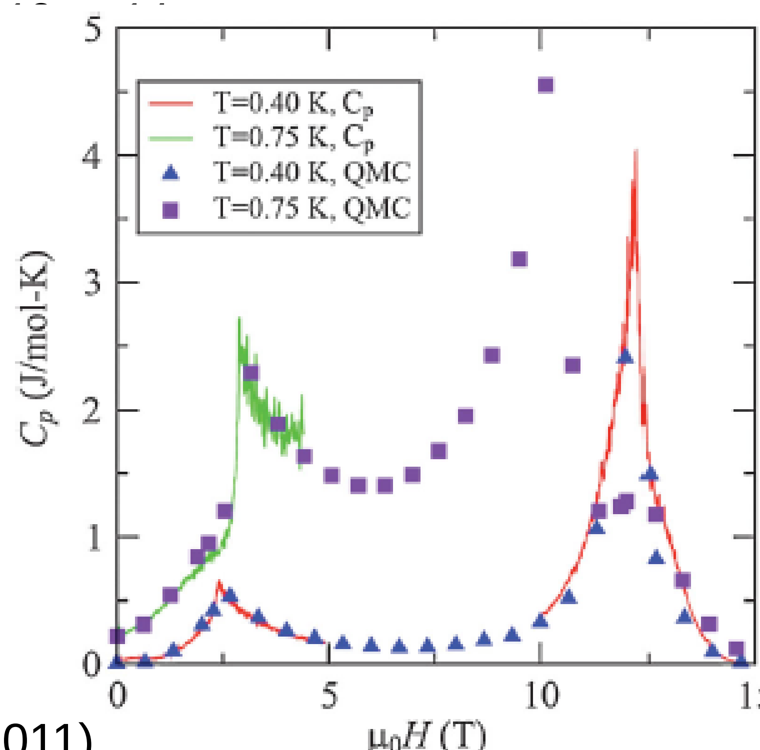
Asymmetry due to quantum fluctuations near H_{c1} but not H_{c2}



$$\frac{1}{m^*} = \frac{\partial^2 \omega}{\partial k^2}$$

Boson mass m* renormalized by quantum fluctuations

$$\frac{m}{m^*} \approx \frac{H_{c2}}{4H_{c1}}\left(1 + \sqrt{1 + \frac{8H_{c1}^2}{H_{c2}^2}}\right)$$



Y. Kohama et al, *Phys. Rev. Lett.* **106**, 037203 (2011).

Beyond Bose-Einstein Condensation

- **Bose Glasses**

  Conducting waves of bosons interacting with random potentials create localization

  (Similar to Anderson localization)

- **Supersolids?**

  Simultaneous diagonal (Ising-like) order and off-diagonal (BEC) order

Bose Glass to BEC transition

Small amount of disorder leads to local disconnected pockets of BEC

Valid in the dilute limit of bosons

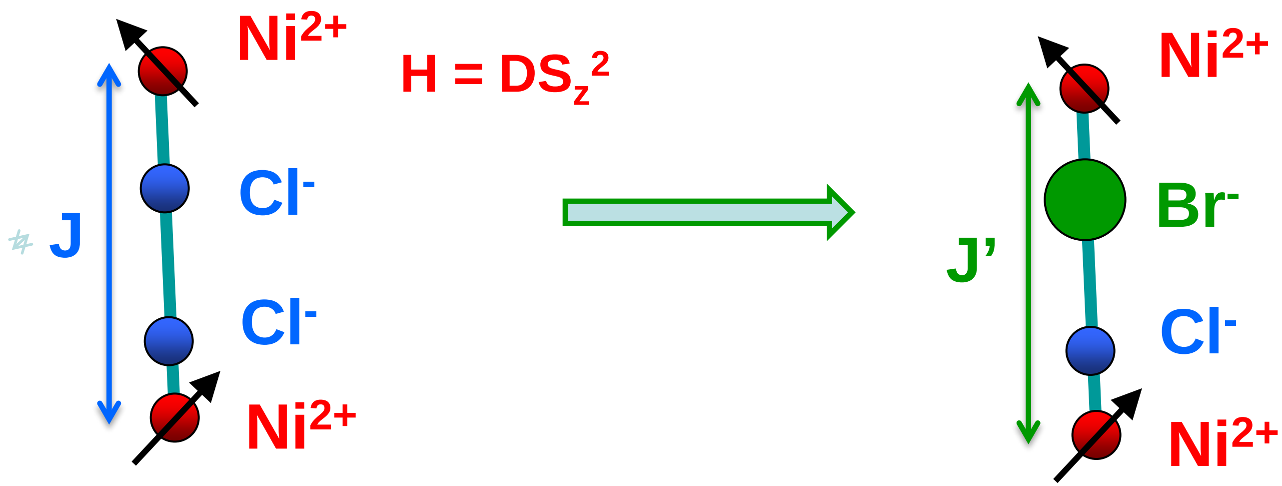
Analogous to Anderson localization (Bloch waves localize due to self-interference when scattering off random potentials)

Sought after in ^{3}He adsorbed on random surfaces
Atomic BECs in random potentials
High-Tc superconductors
Photonic systems
Others

Creating a Bose Glass:
Introduce disorder via Br substitution

8% Br substitution

$NiCl_{2-x}Br_x$-$4SC(NH_2)_2$



J -> J' = 2.35J

D -> D' = D/2

A rare example of clean doping:

X-rays support no change in lattice parameter and no buckling of structure
Only one Cl site (the larger one) supports Br substitution

R. Yu et al, *Nature*, submitted

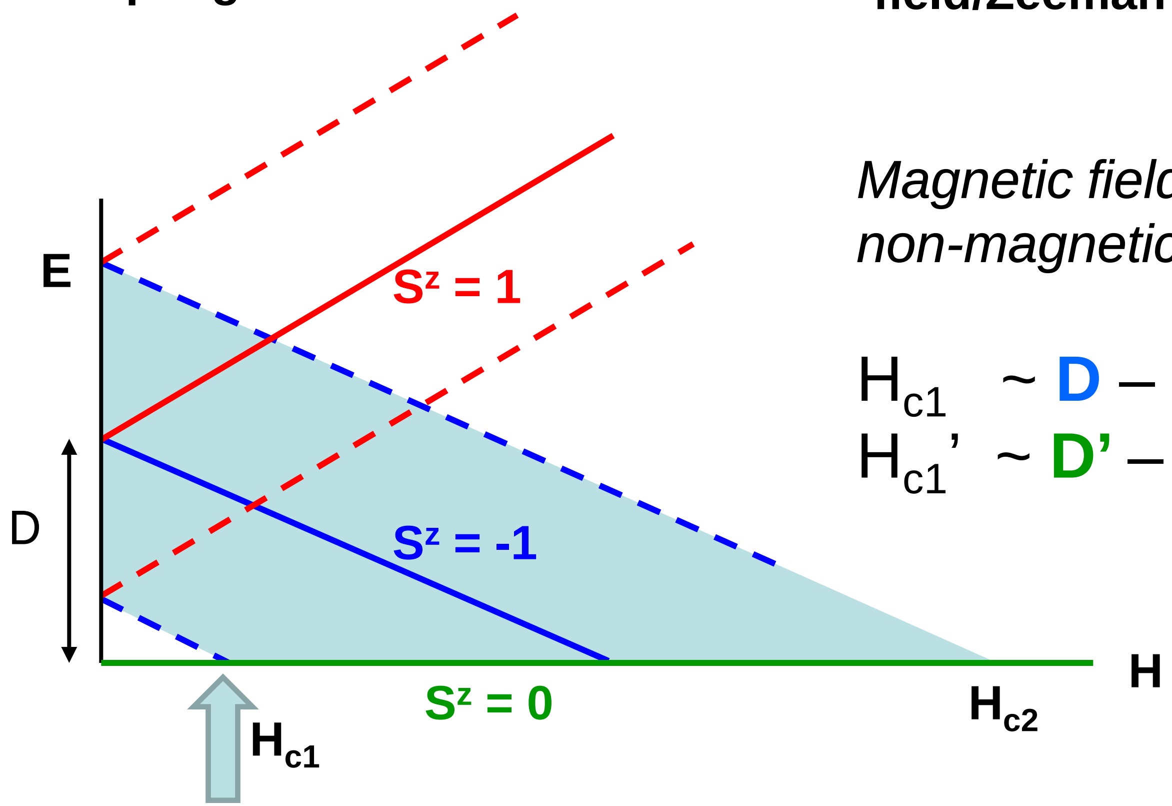
Bose-Einstein Condensation

$$\mathrm{H} = D\sum_{i}\left(S_i^z\right)^2 + \sum_{<i,j>\nu} J_\nu \vec{S}_i \cdot \vec{S}_j - g\mu_B H \sum_i S_i^z$$

Spin-orbit coupling

AFM exchange

Magnetic field/Zeeman term



Magnetic field to overcome non-magnetic ground state

$H_{c1} \sim D - 4zJ$ Ni-Cl-Cl-Ni

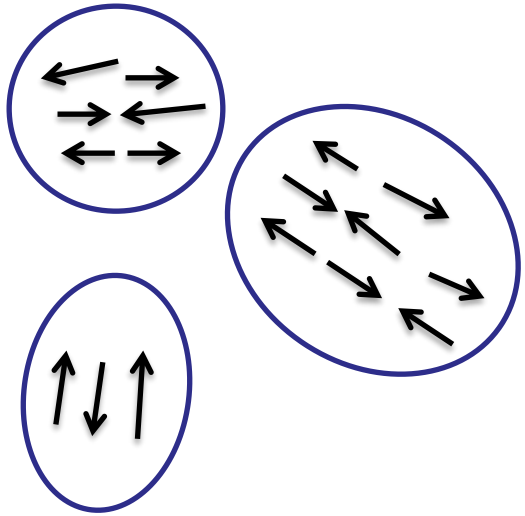
$H_{c1}' \sim D' - 4zJ'$ Ni-Br-Cl-Ni

$H < H_{c1'}$

All Ni in $S_z = 0$ state
No ordering
No magnetization

$S_z = 0$

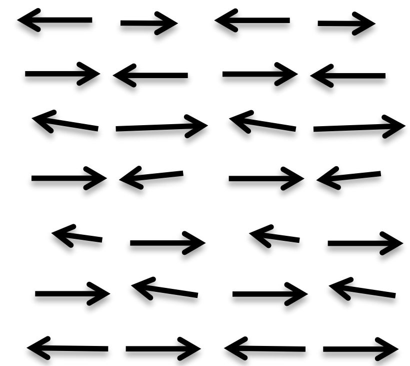
$H_{c1'} < H < H_{c1}$



Clusters of Ni connected by Br start to order
(In reality these are 3-D)

Bose Glass

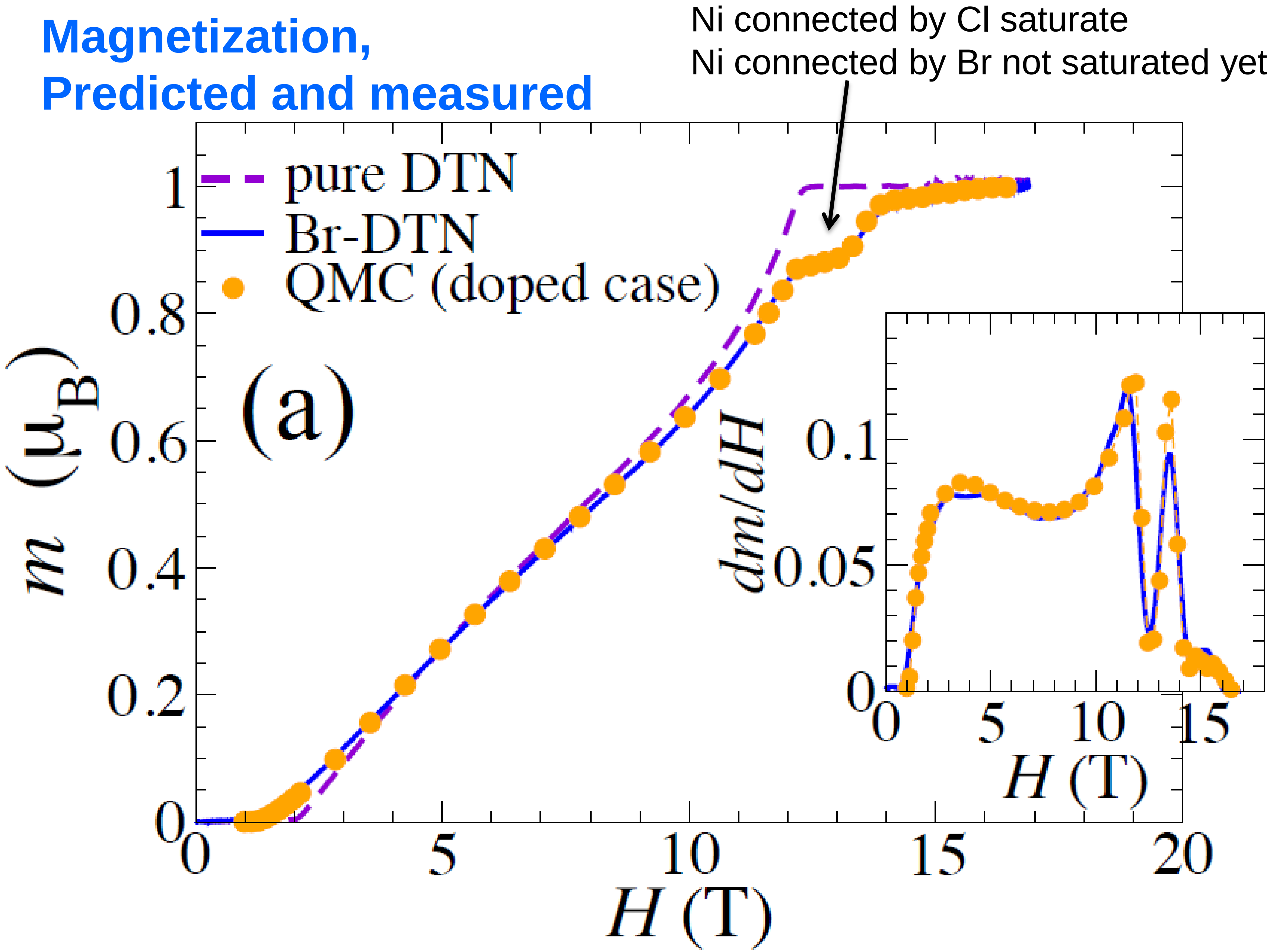
$H_{c1} < H$



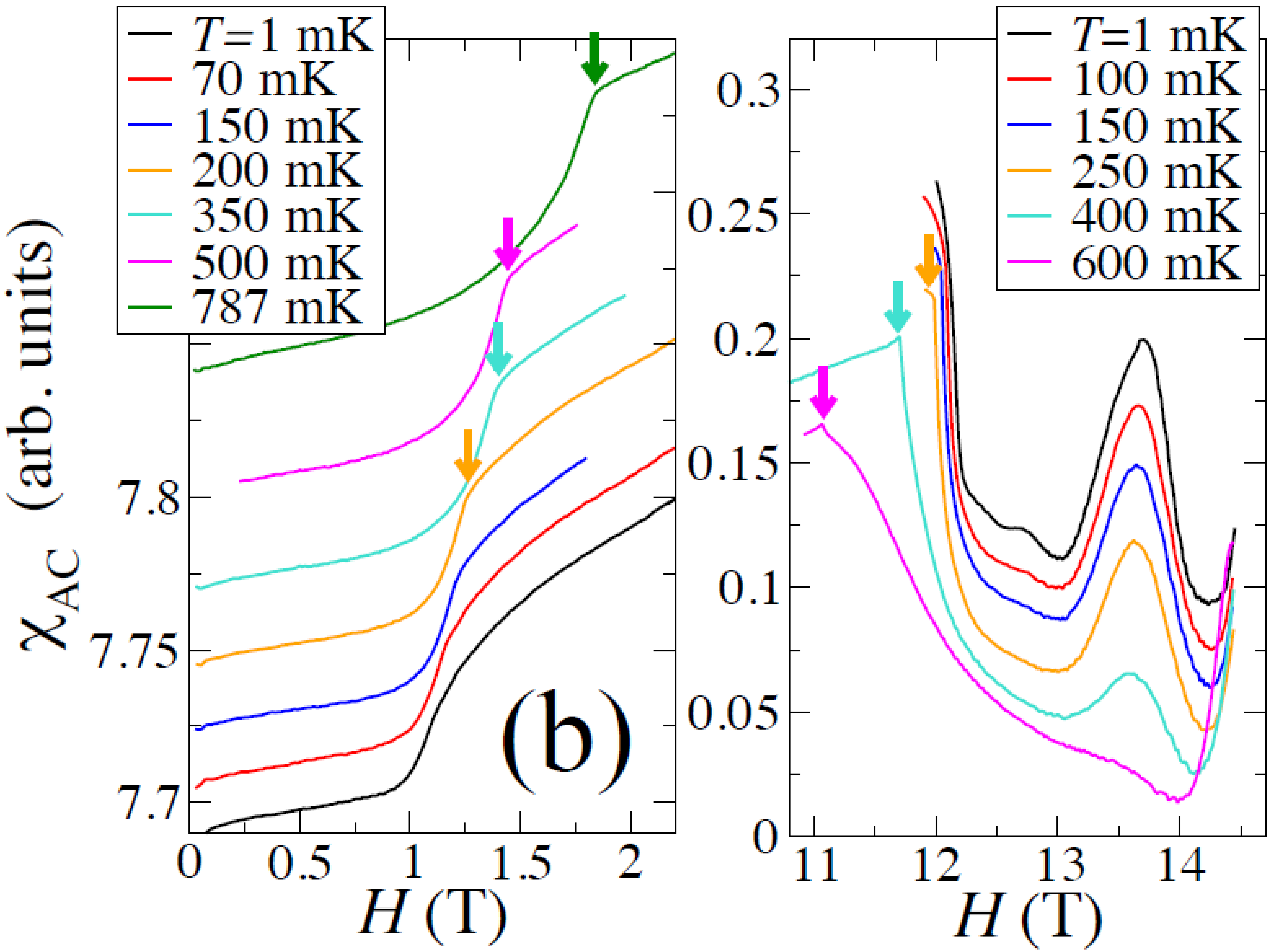
Br and Cl-coupled spins order

Inhomogenous BEC

Magnetization, Predicted and measured

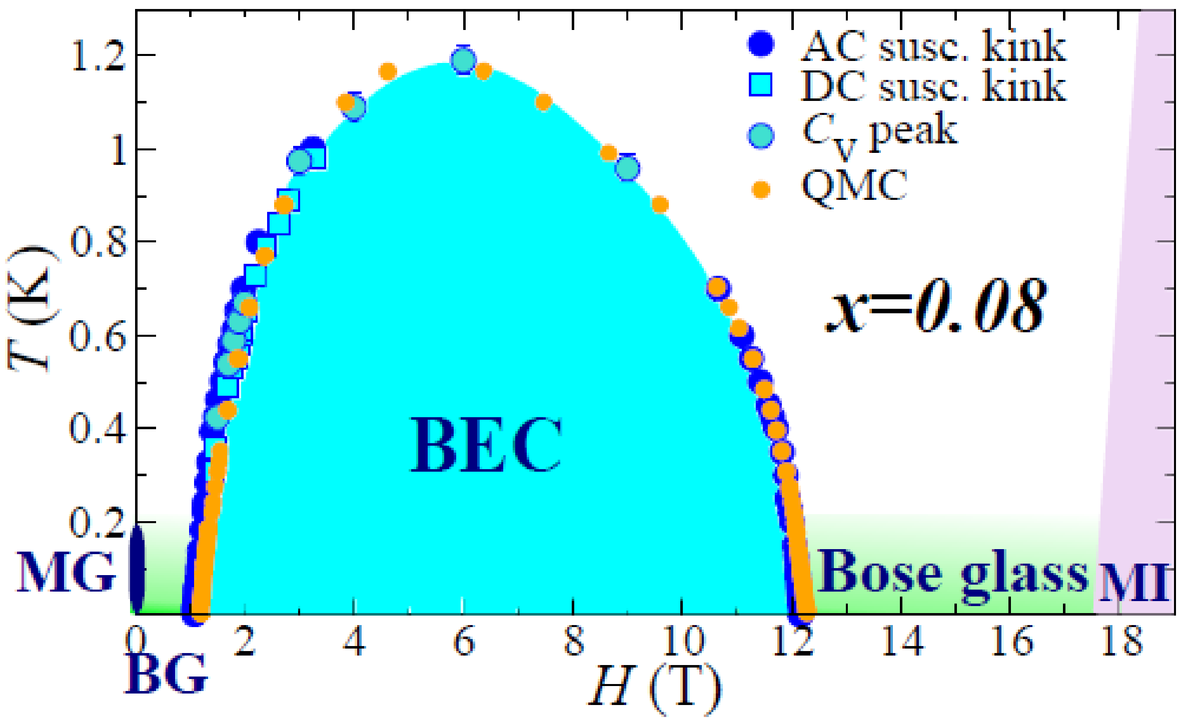


Ac Susceptibility to determine phase diagram

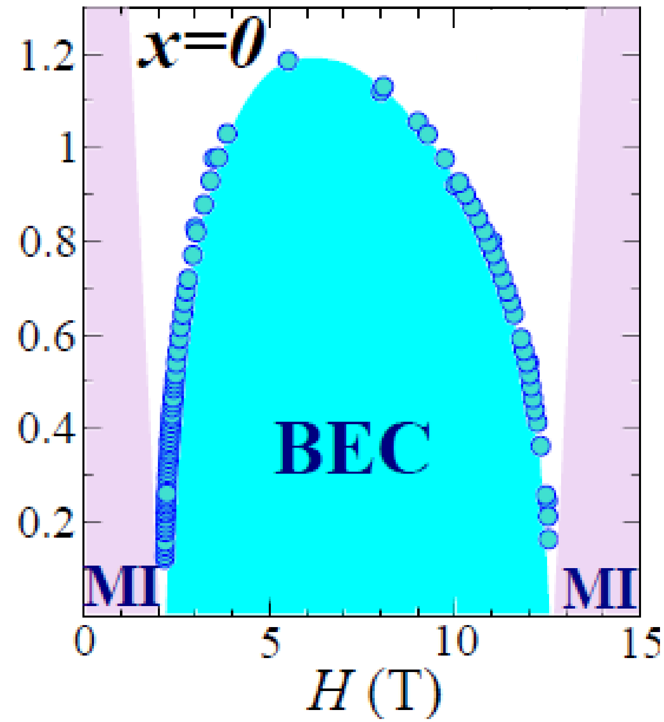


Experimental phase diagrams

Br-Doped DTN



Pure DTN



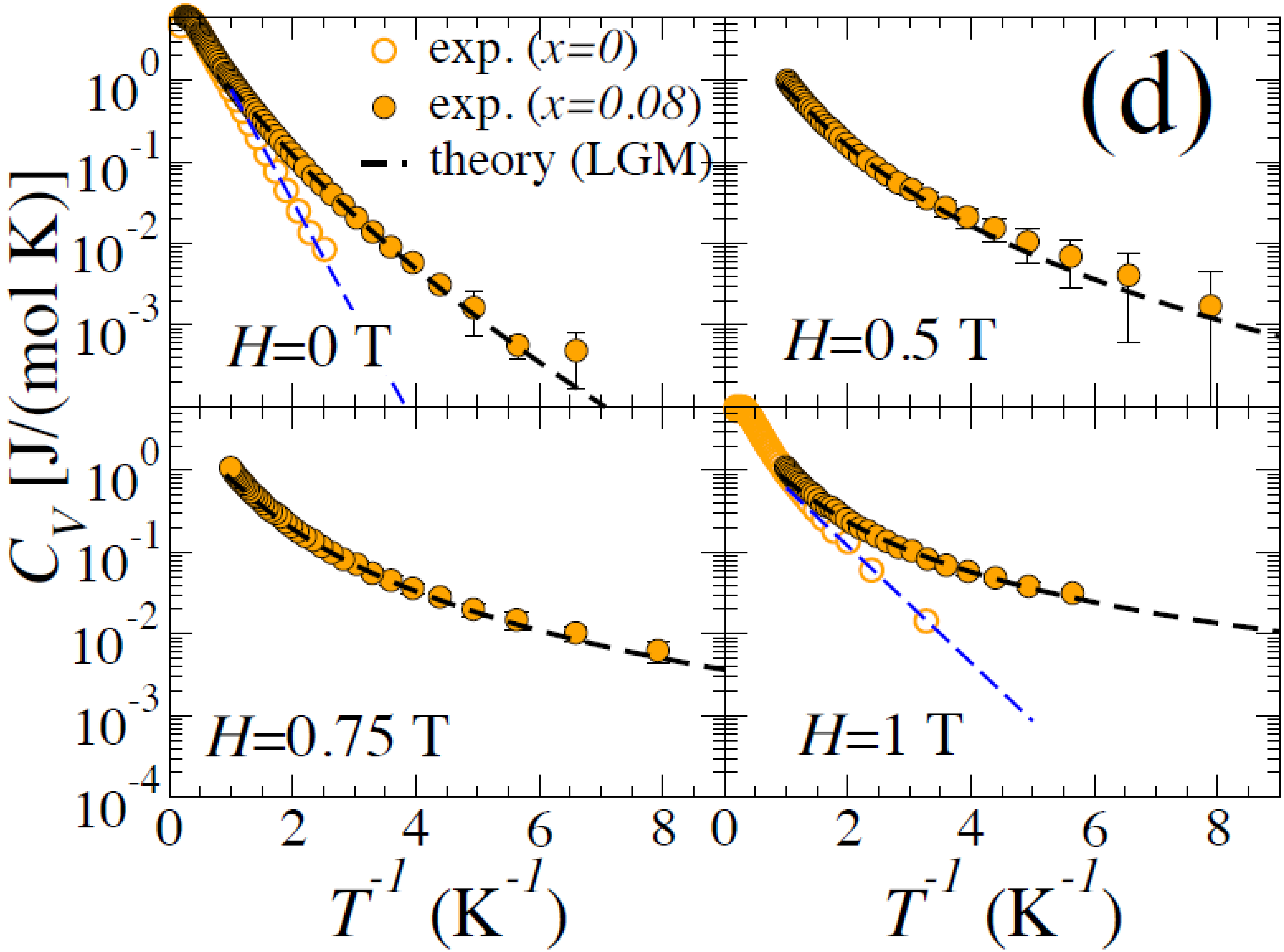
MI – Mott Insulator

MG – Mott Glass

BG – Bose Glass

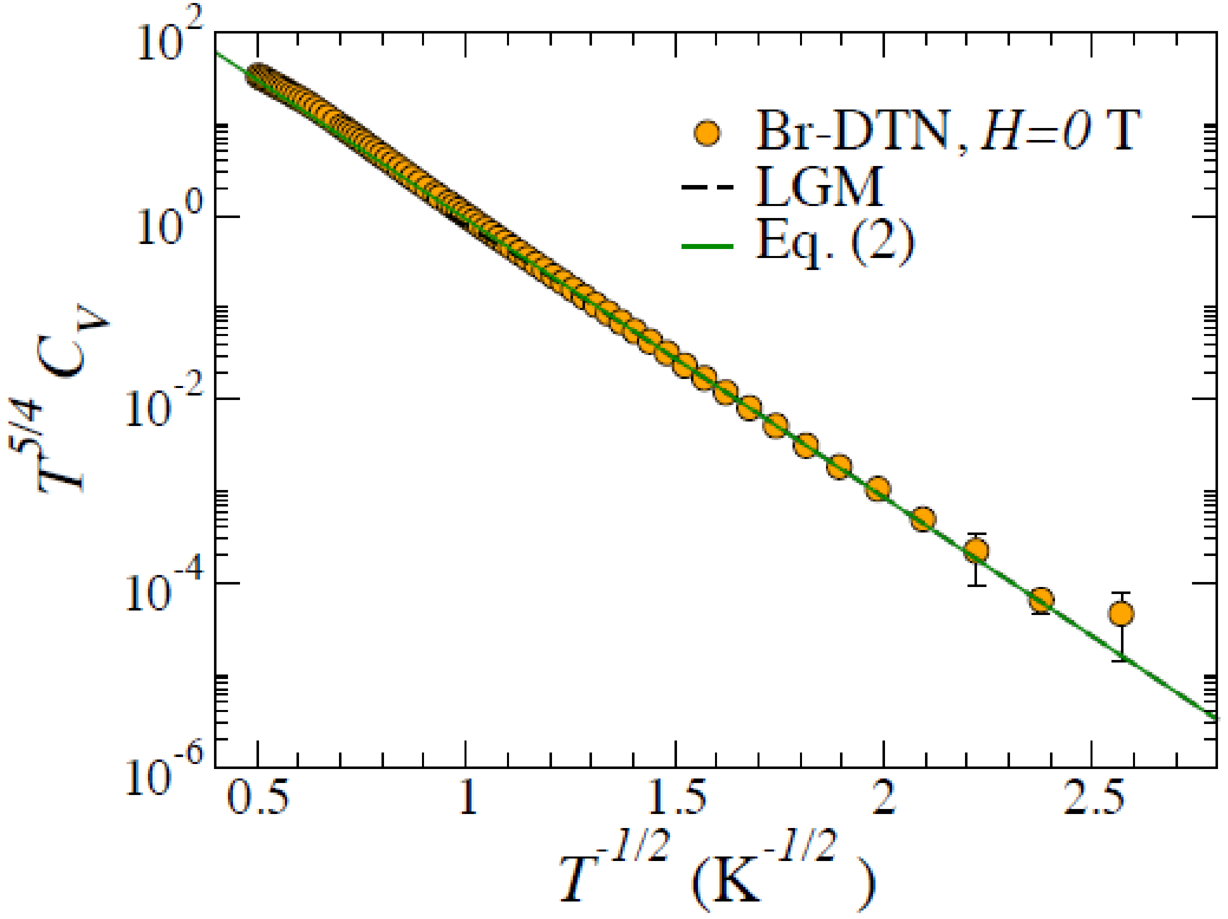
BEC – Bose-Einstein Condensate

Specific heat, predicted and measured

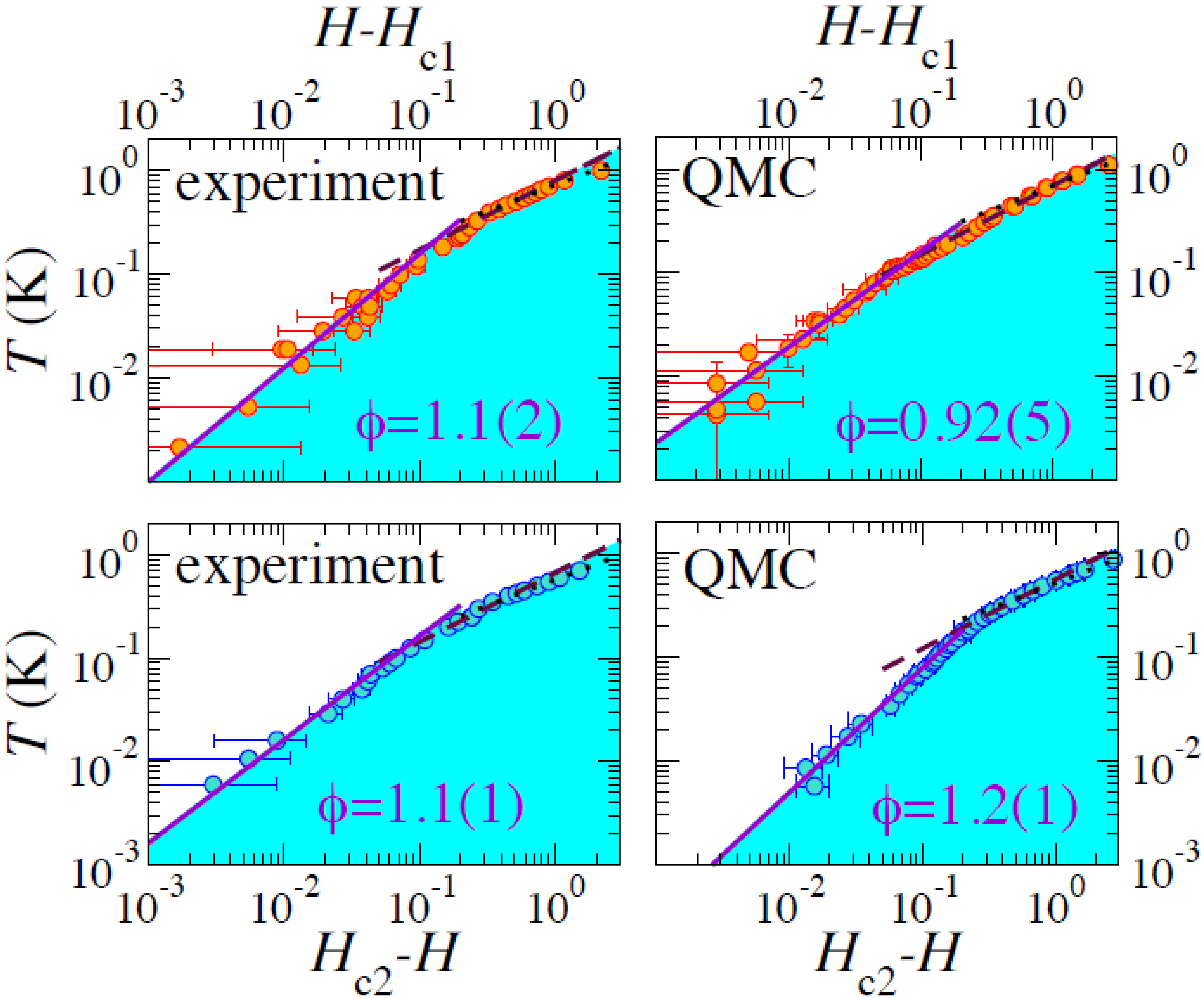


Possibly a Mott Glass at zero magnetic field

- If so, first experimental realization
- Vanishing magnetization at zero field
- Yet the system is globally gapless to spin excitations
  [Gap D-4zJ depends on size of Br-connected cluster
  Statistically there is no upper limit to the size of Br clusters]



Predicted and measured power-laws of the critical fields between BEC and BG



Acknowledgements

Crystal growth and magnetization
Armando Paduan-Filho
Universidade de Sao Paulo, Brazil

NHMFL-LANL
Marcelo Jaime, Diego Zocco, Yoshi Kohama, Alex Lacerda

1 mK (High B/T laboratory, U. Florida)
Liang Yin, Jian-sheng Xia, Neil Sullivan

Theory
Tommaso Roscilde *U. Lyons*
Cristian Batista, *LANL*
Sasha Chernychev*, UC Irvine*

Magnetostriction
Victor Correa, Stan Tozer
NHMFL-Tallahassee

Resonant Ultrasound
Cristian Pantea, Jon Betts, Albert Migliori, *NHMFL-LANL*

ESR
Sergei Zvyagin, *Dresden Hochfeld*
Steve Hill, *NHMFL*
Ross McDonald, *NHMFL*

Thermal conductivity
Alex Solugobenko, John Mydosh (*formerly U. Cologne*)
Franziska Weickert *LANL*

Quantum Monte Carlo
Stephan Haas *USC*
Pinaki Sengupta *LANL*
Rong Yu *Rice*

NHMFL-Tallahassee
Tim Murphy, Eric Palm

NSF NHMFL DOE

Key papers

R. Yu et al, *Nature*, in revision, arXiv:cond-mat1109.4403v2 (Bose Glass)

Y. Kohama et al, *Phys. Rev. Lett* **106**, 037203 (2011). (Quantum fluct. renorm. boson mass)

L. Yin et al, *Phys. Rev. Lett* **101**, 187205 (2008). (BEC shown to 1 mK)

S. Zvyagin et al, *Phys. Rev. Lett* **98***,* 047205 (2007). (ESR measurements)

V. S. Zapf et al, *Phys. Rev. Lett* **96**, 077204 (2006). (First paper showing BEC)

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