DISORDER-INDUCED REVIVAL OF BOSE-EINSTEIN CONDENSATION IN $\mathrm{Ni(Cl_{1-x}Br_x)_2 - 4SC(NH_2)}$ AT HIGH MAGNETIC FIELDS

Nicolas Laflorencie

CNRS - LPT Toulouse

OUTLINE

Impurity-doped spin gapped systems at zero magnetic field

- A generic impurity-induced ordering mechanism

Finite magnetic field

- Field-induced and disorder-induced phase transitions?
- The DTN(X) material: coupled (disordered) S=1 chains
- A nice example of a subtle competition between delocalization (BEC) and localization (Bose Glass)

OUTLINE

Impurity-doped spin gapped systems at zero magnetic field

BCPO
$\mathrm{Bi(Cu_{1-p}(Zn\ or\ Ni)_p)_2PO_6}$

- Bobroff *et al.* PRL 2009
- Alexander *et al.* PRB 2010

Julien Bobroff
Philippe Mendels

NMR + μSR

Finite magnetic field

DTNX
$\mathrm{Ni(Cl_{1-x}Br_x)_2 - 4SC(NH_2)_2}$

- Blinder *et al.* PRB(R) 2017
- Dupont *et al.* PRL 2017
- Orlova *et al.* PRL 2017
- Dupont *et al.* PRB 2017
- Orlova *et al.* PRL 2018

Synthesis

Instituto de Física
Universidade de São Paulo

Armando Paduan-Filho

LABORATOIRE DE PHYSIQUE THÉORIQUE TOULOUSE

Maxime Dupont
Sylvain Capponi
Nicolas Laflorencie

Theory

LNCMI

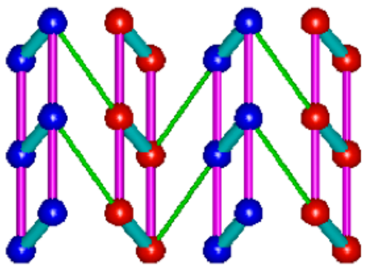
Mladen Horvatić
Claude Berthier (1946-2018)
Hadrien Mayaffre
Edwin Kermarrec
Anna Orlova
Rémi Blinder

NMR

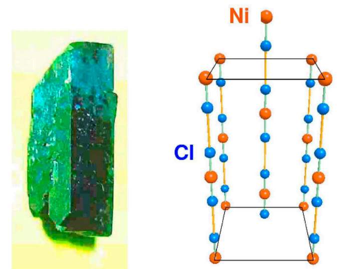
SPIN-GAPPED MATERIALS

- Quasi-1D: Ladders, Haldane chains, spin-Peierls chains...

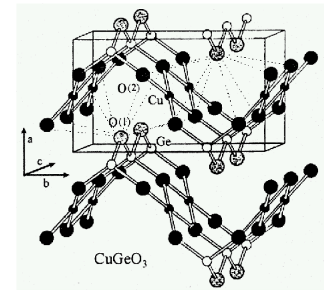
BPCB



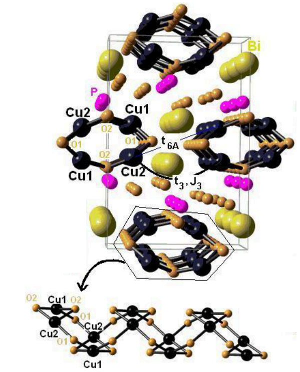
DTN



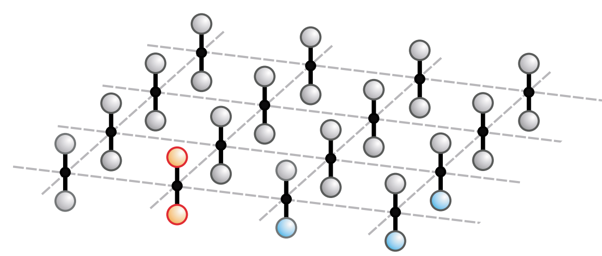
$CuGeO_3$

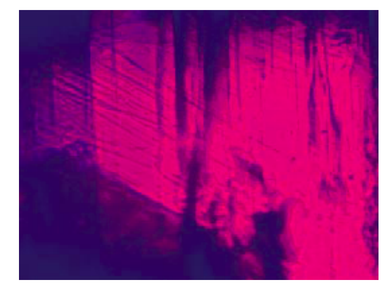


$BiCu_2PO_6$



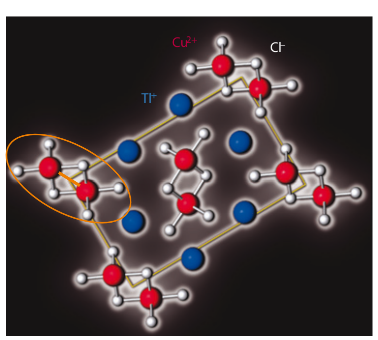
- Quasi-2D: bilayers





$BaCuSi_2O_6$

- 3D coupled dimer systems



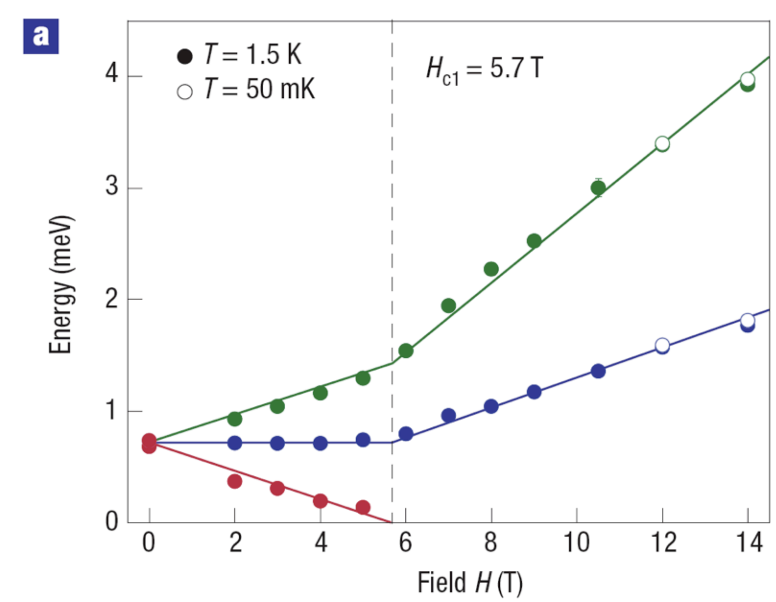
$TlCuCl_3$

Bose–Einstein condensation of the triplet states in the magnetic insulator $TlCuCl_3$

Ch. Rüegg*, N. Cavadini*, A. Furrer*, H.-U. Güdel†, K. Krämer†, H. Mutka‡, A. Wildes‡, K. Habicht§ & P. Vorderwisch§

* *Laboratory for Neutron Scattering, ETH Zürich & Paul Scherrer Institut, 5232 Villigen PSI, Switzerland*
† *Department of Chemistry and Biochemistry, Universität Bern, 3000 Bern 9, Switzerland*
‡ *Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France*
§ *BENSC, Hahn-Meitner-Institut, 14109 Berlin Wannsee, Germany*

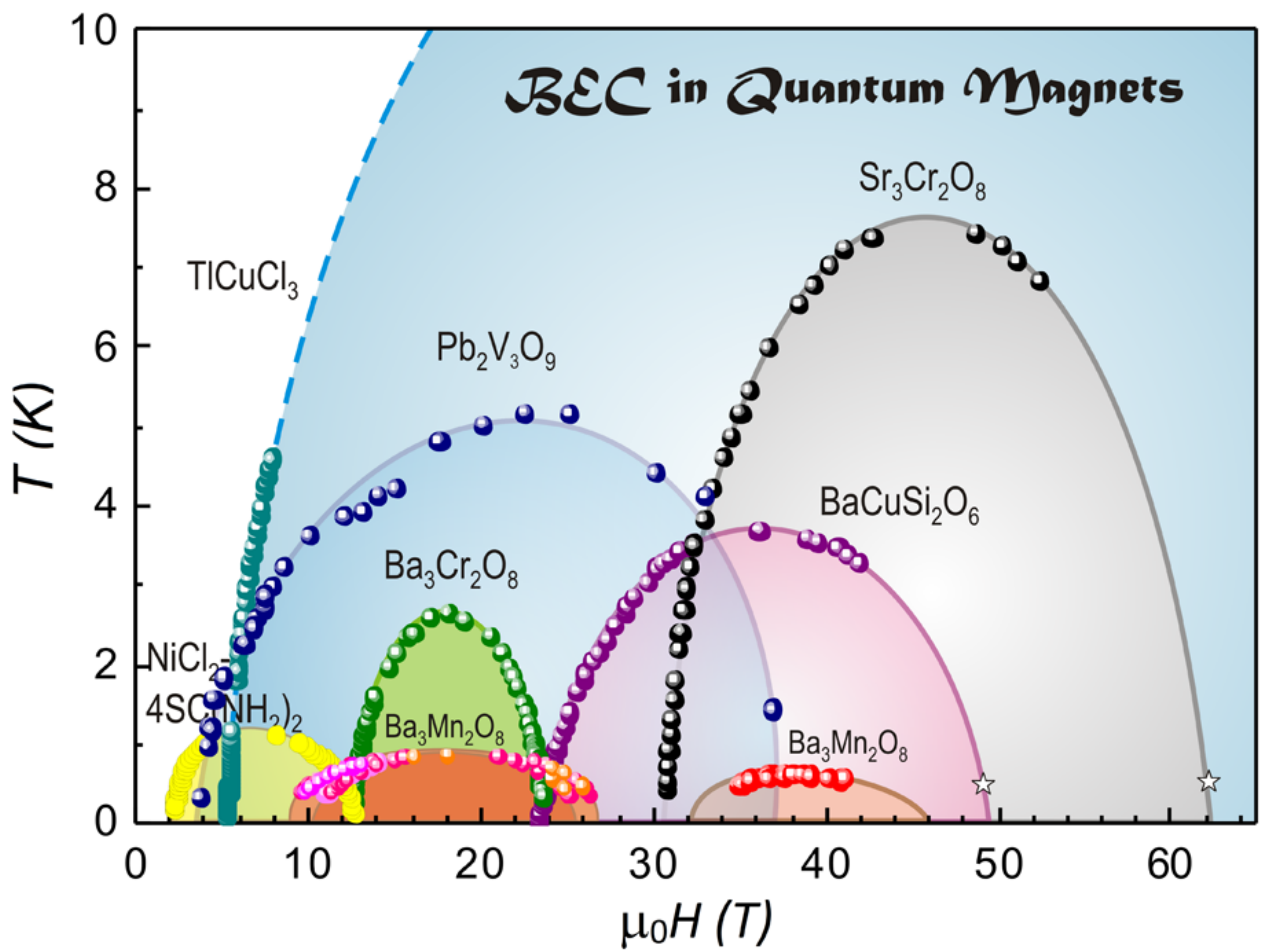
Rüegg *et al.* Nature (2003)



MAGNETIC FIELD INDUCED BOSE-EINSTEIN CONDENSATION

T. Giamarchi, C. Rüegg, O. Tchernyshyov
Nature Physics 2008

V. Zapf, M. Jaime and C. D. Batista
Rev. Mod. Phys. 2014

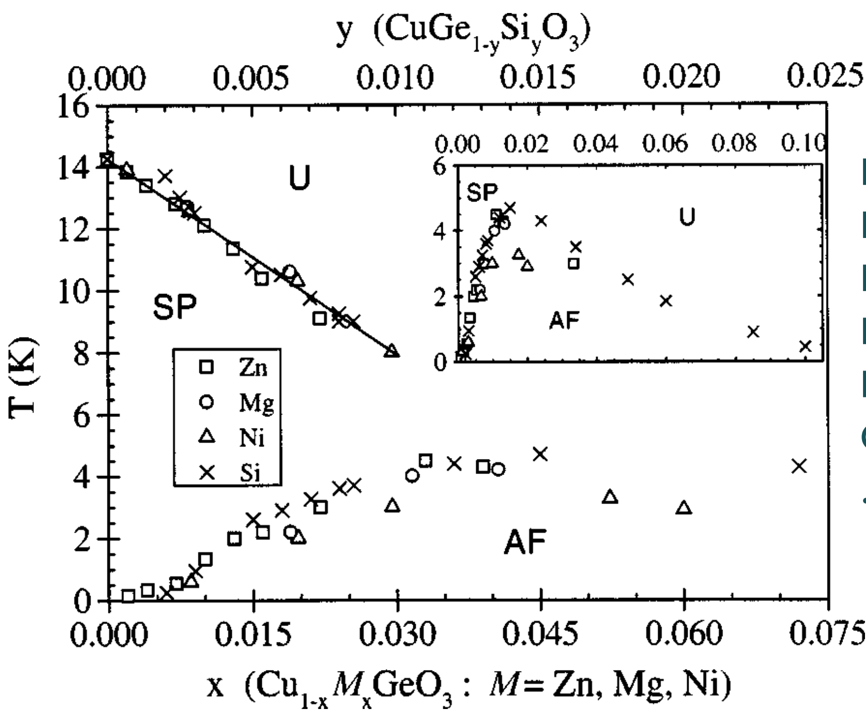


ZERO MAGNETIC FIELD: A GENERIC IMPURITY-INDUCED ORDERING

- Spin-Peierls
- 2-leg Ladders
- Coupled dimers
- Coupled Haldane chains

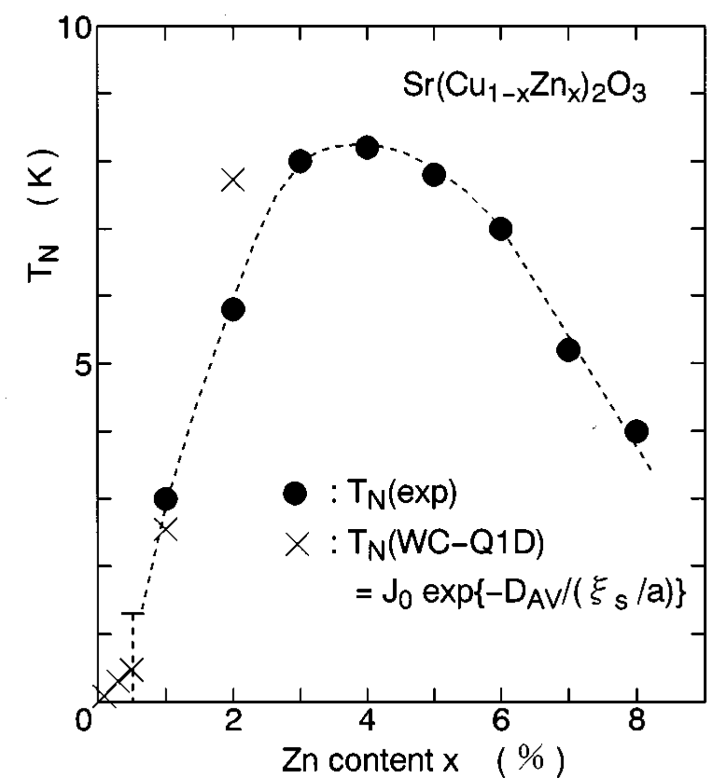
ZERO MAGNETIC FIELD: A GENERIC IMPURITY-INDUCED ORDERING

◉ Spin-Peierls



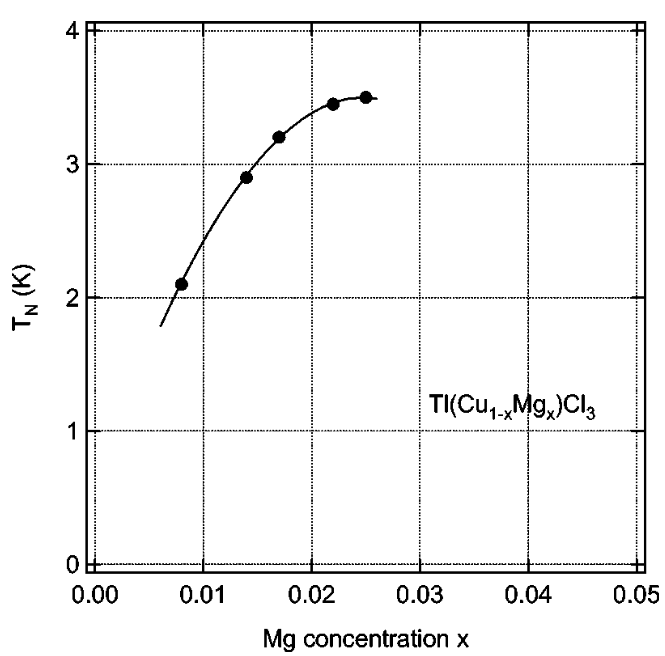
Hase *et al.* PRL 1993
Regnault *et al.* EPL 1995
Fukuyama *et al.* JPSJ 1996
Martin *et al.* PRB 1995
Masuda *et al.* PRL 1998
Grenier *et al.* PRB 1998
…

◉ 2-leg Ladders



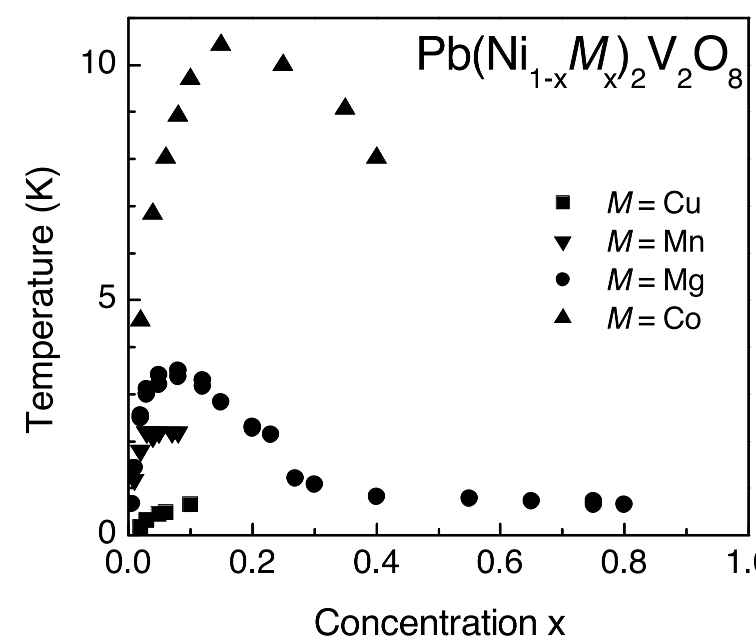
Azuma *et al.* PRB 1997
Greven *et al.* PRL 1998
Ogushi *et al.* PRB 1999
Larkin *et al.* PRL 2000
…

◉ Coupled dimers



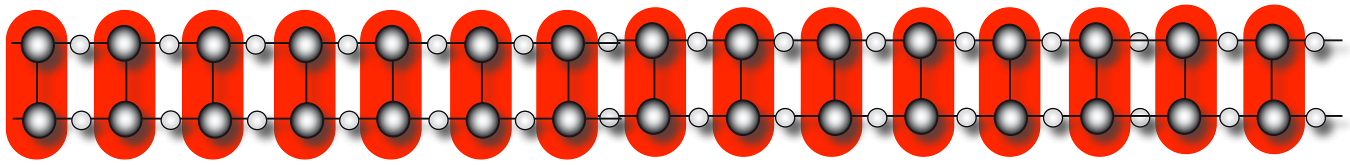
Oosawa *et al.* PRB 2002
Oosawa *et al.* PRB 2003
Fujisawa *et al.* PRB 2006
Suzuki *et al.* PRB 2011
…

◉ Coupled Haldane chains



Uchiyama *et al.* PRL 1999
Smirnov *et al.* PRB 2002
Imai *et al.* 2004
Arčon *et al.* EPL 2004
Zorko *et al.* PRB 2006
…

SINGLE IMPURITY PHYSICS



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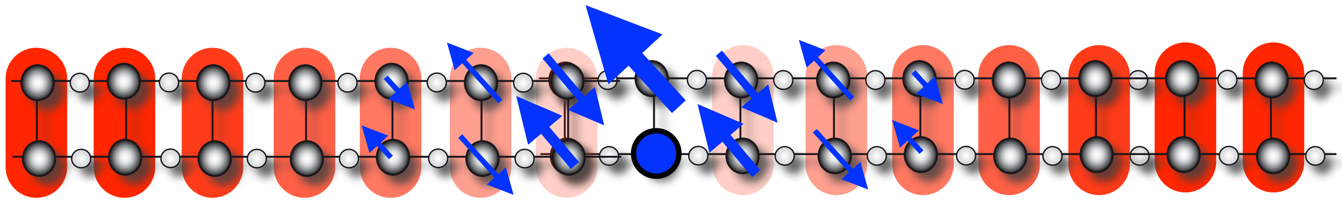
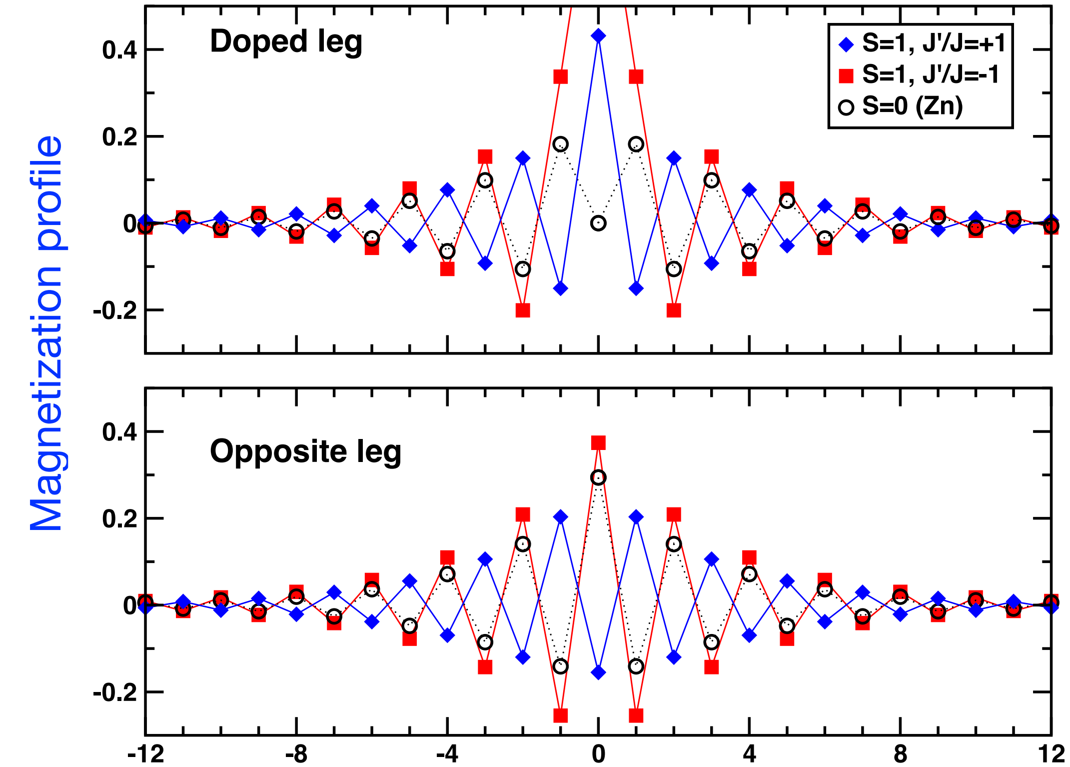


$\xi \simeq J/\Delta$

- A non-magnetic impurity breaks a singlet and releases a spin S=1/2, confined in the vicinity of the impurity

$$\langle S_r^z \rangle \sim (-1)^r \exp(-r/\xi)$$



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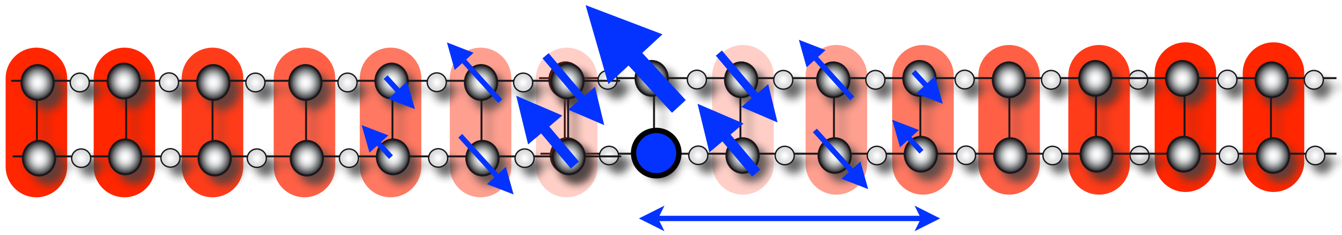

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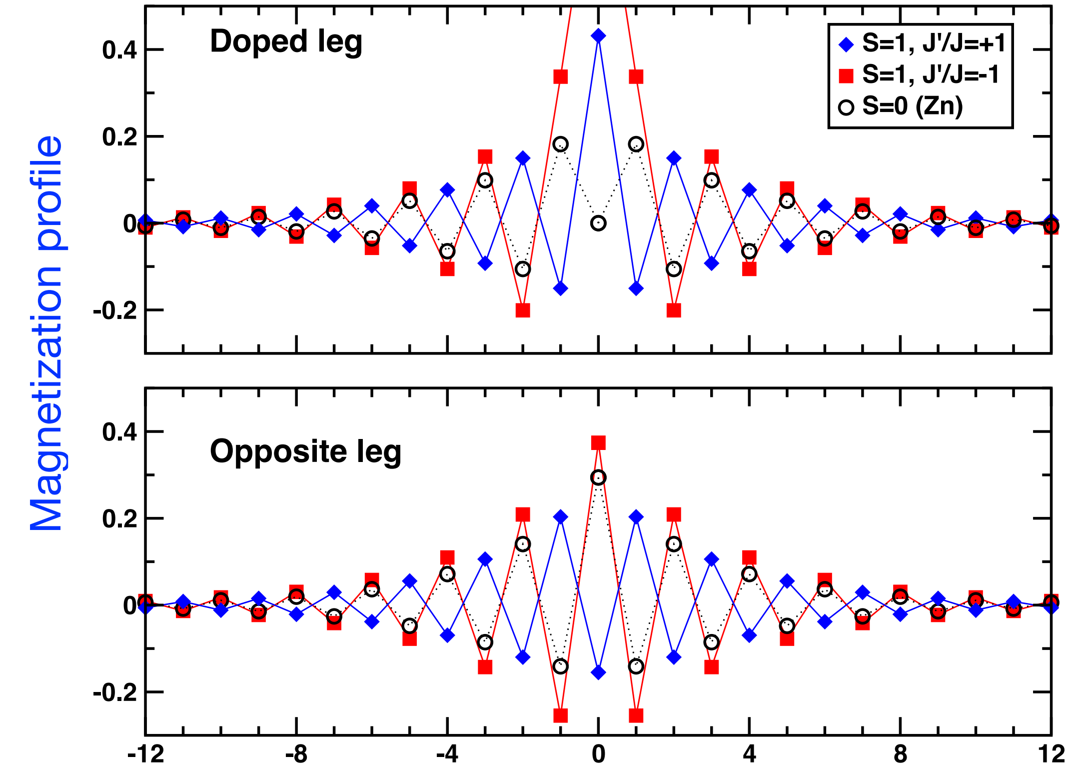
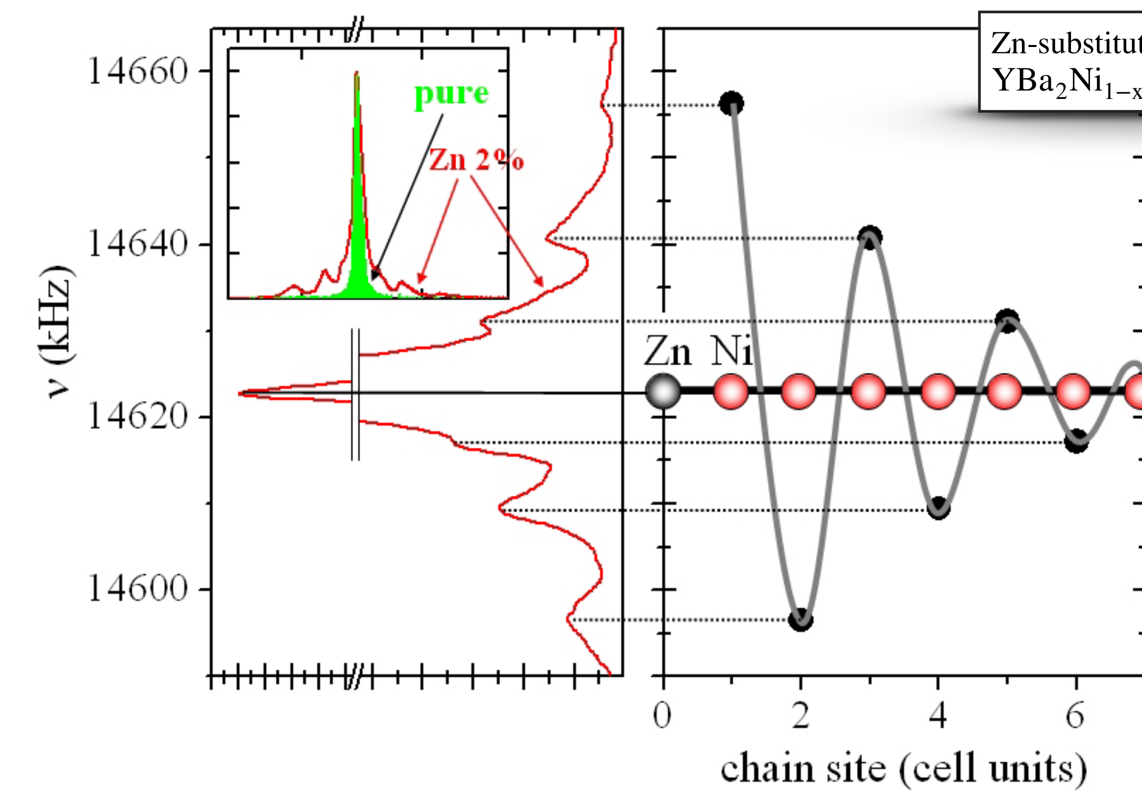


- Clearly visible in NMR spectra



Zn-substituted Haldane chains $YBa_2Ni_{1-x}Zn_xO_5$.

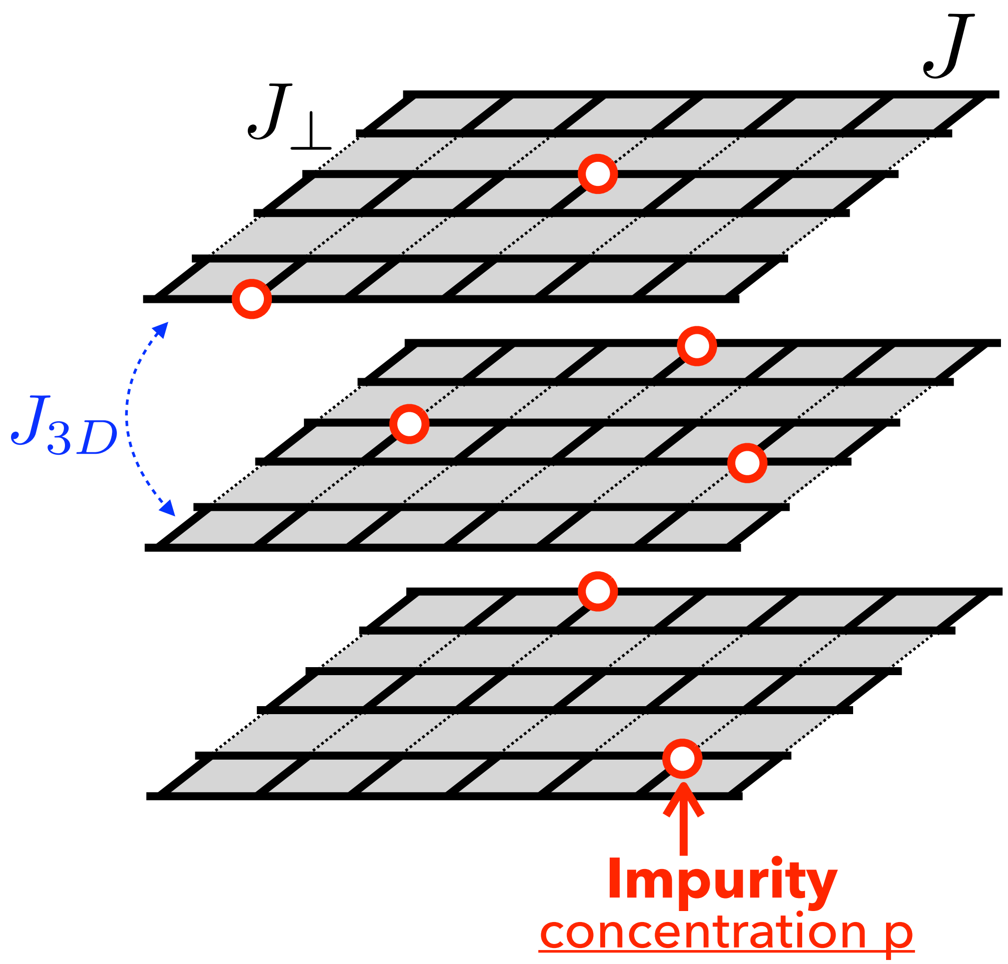
See: Tedoldi *et al.* PRL 1999

$\langle S_Z \rangle$

MANY IMPURITIES

J. Bobroff *et al.* PRL 2009

- Real 3D materials



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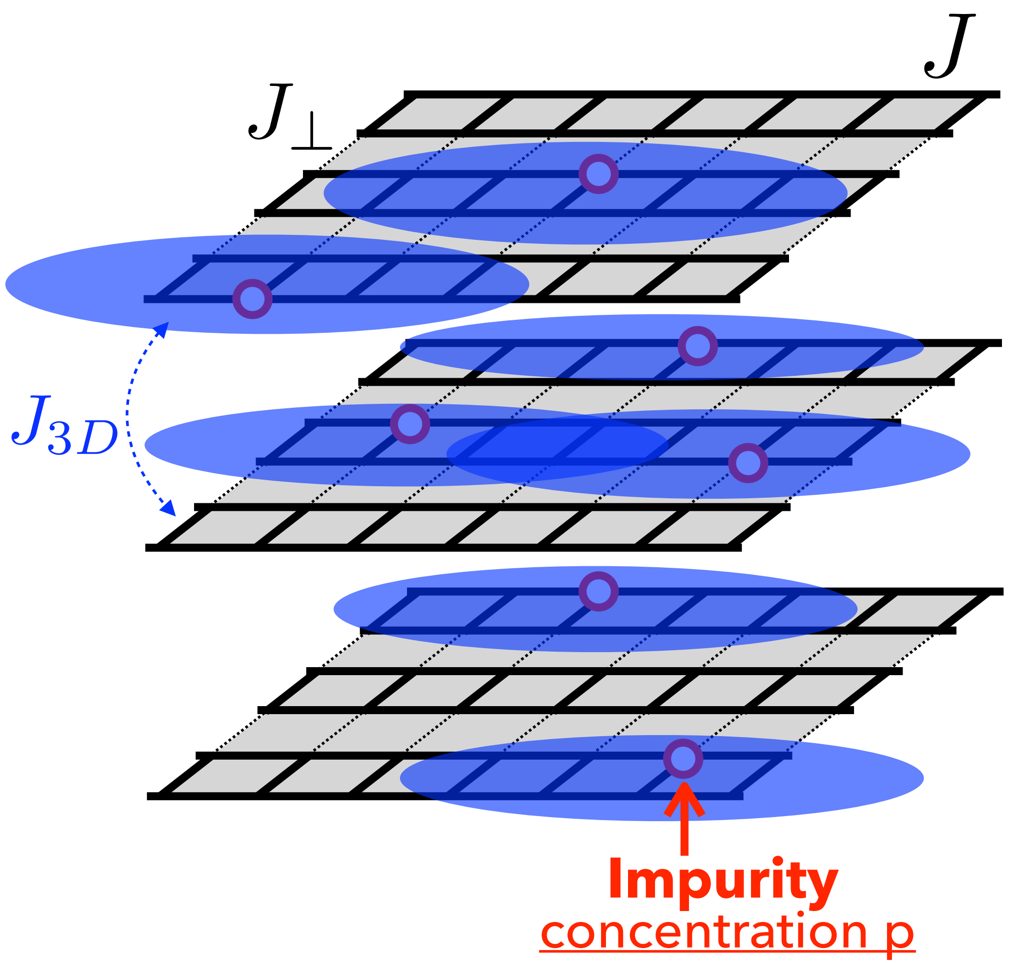

$$V_\xi = \xi_x \xi_y \xi_z$$

Volume of the AF correlated clouds

- Effective interaction

Sigrist and Furusaki JPSJ 1996

$$J^{\mathrm{eff}}(x,y,z) \propto (-1)^{x+y+z} \exp\left(-\frac{x}{\xi_x} - \frac{y}{\xi_y} - \frac{z}{\xi_z}\right)$$

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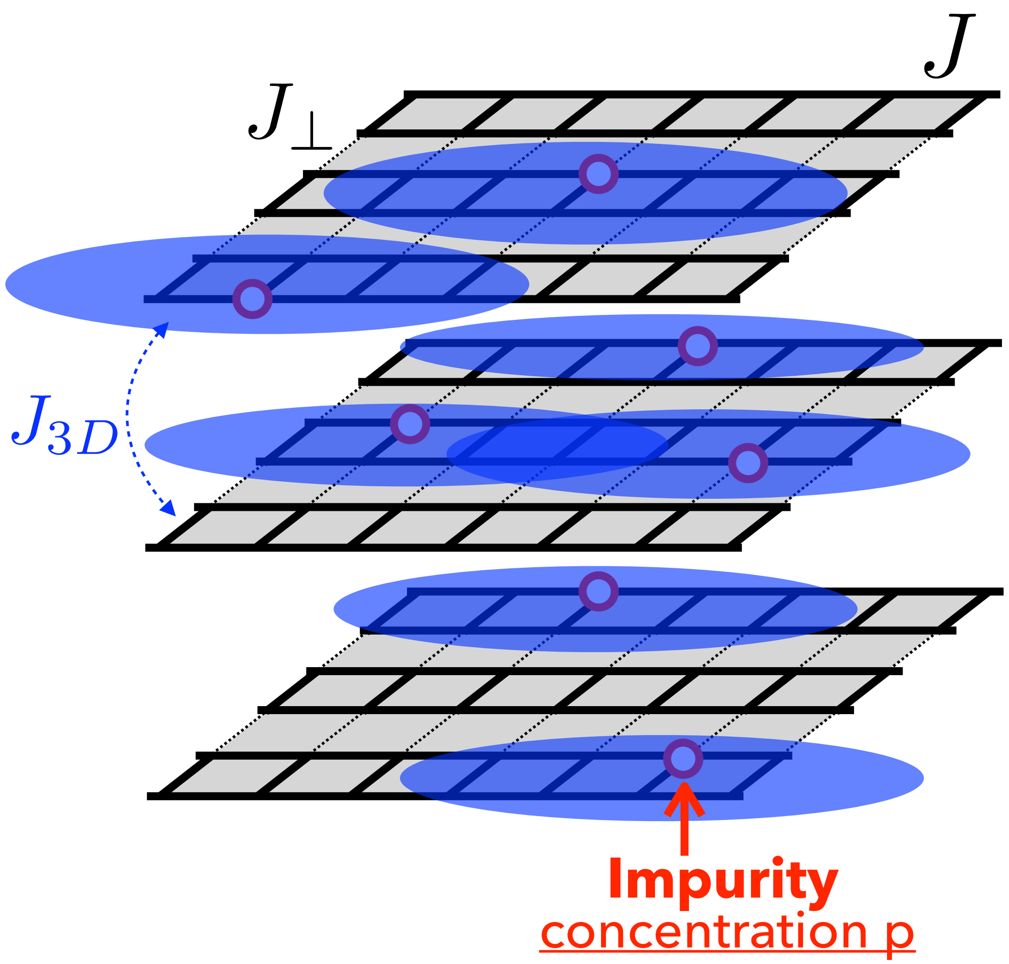

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- 3D ordering controlled by $J_{3D} \exp\left(-\frac{x}{\xi_x} - \frac{y}{\xi_y} - \frac{z}{\xi_z}\right)$

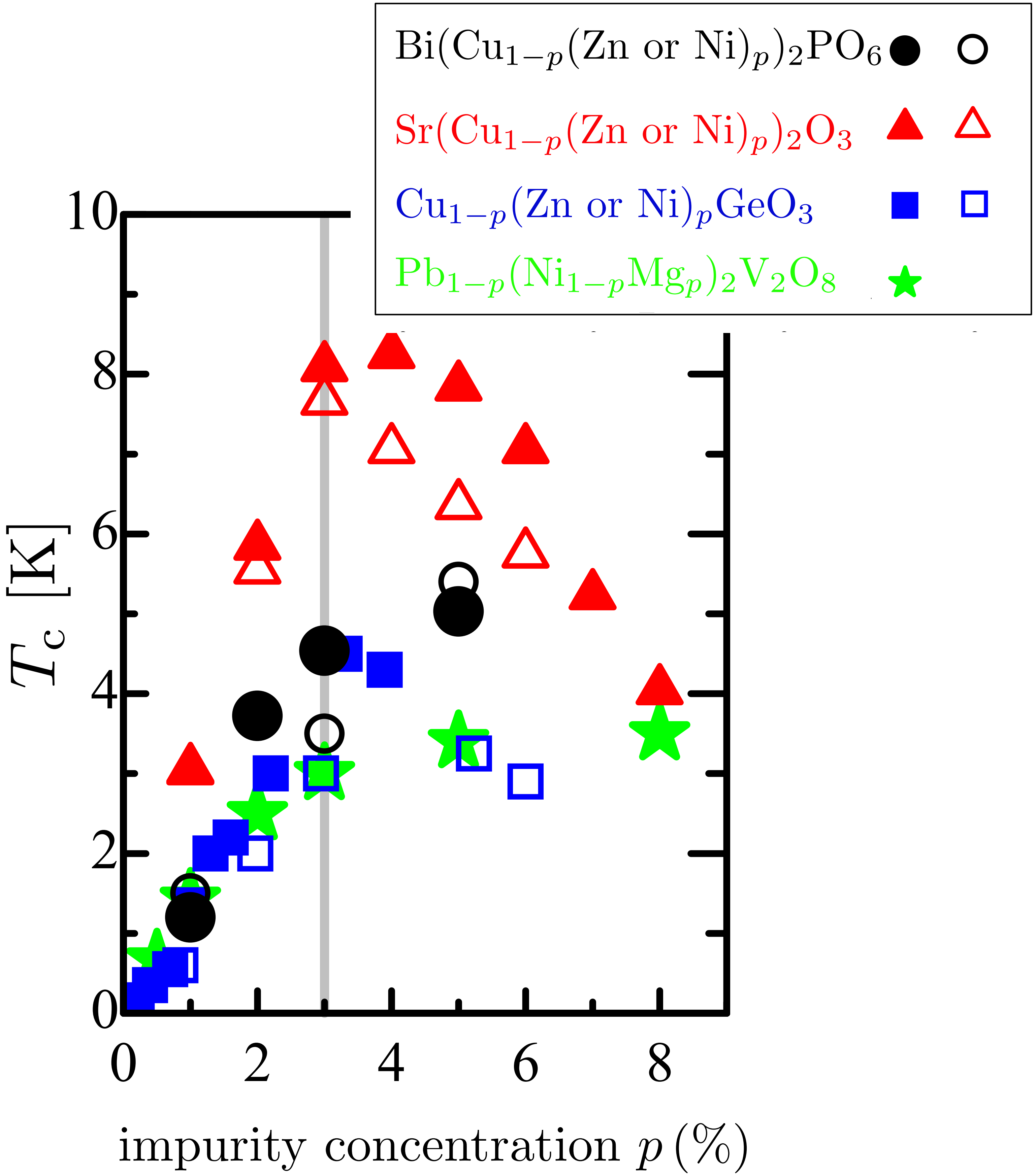
- Average 3D coupling

$$\boxed{T_c \approx \langle J_{3D}^{\text{eff}} \rangle = J_{3D} \times V_\xi \times p}$$

if $V_\xi \, p < 1$

A GENERIC "ORDER-FROM-DISORDER" SCENARIO

J. Villain *et al.* 1980
J. Bobroff *et al.* PRL 2009



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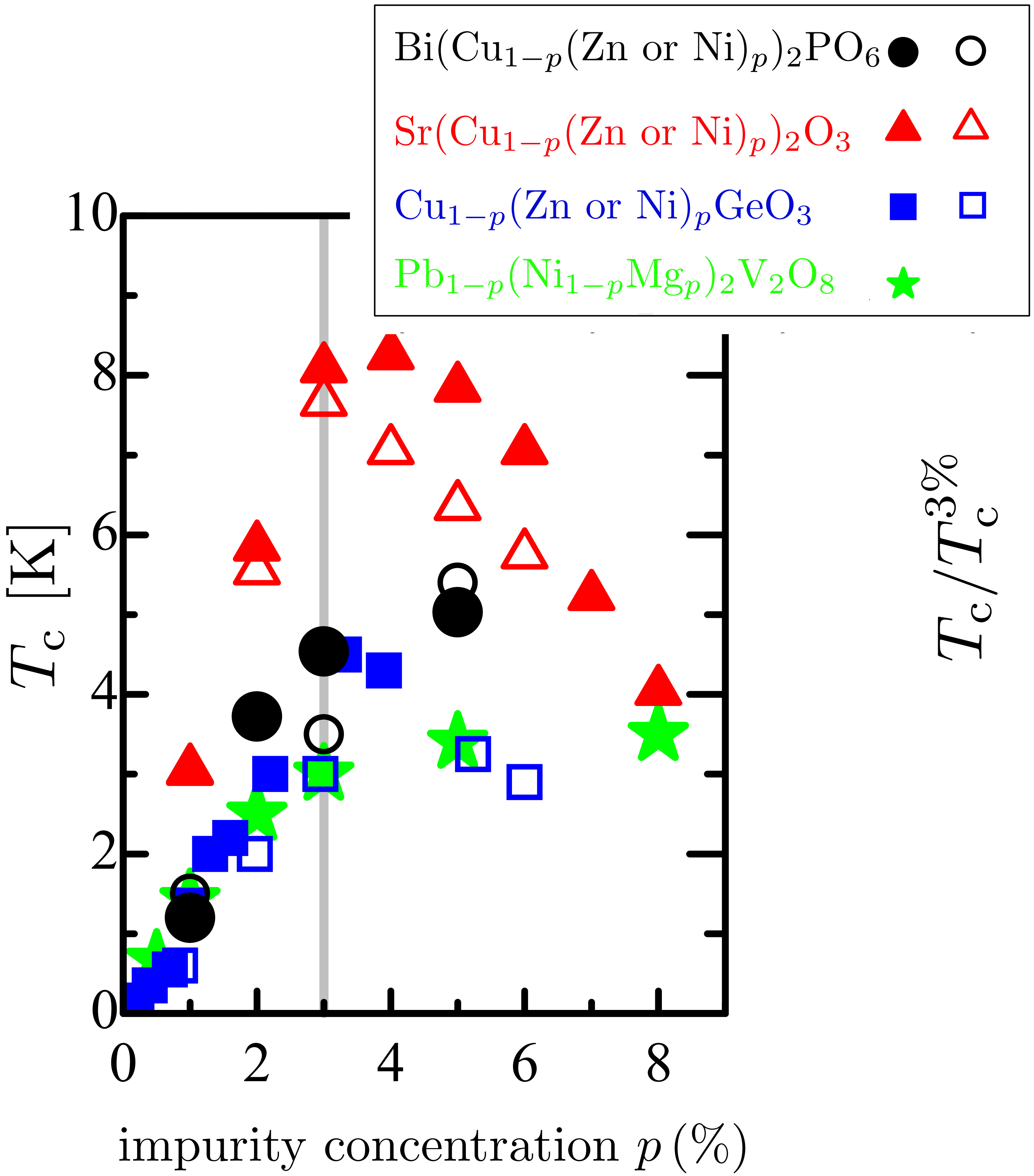

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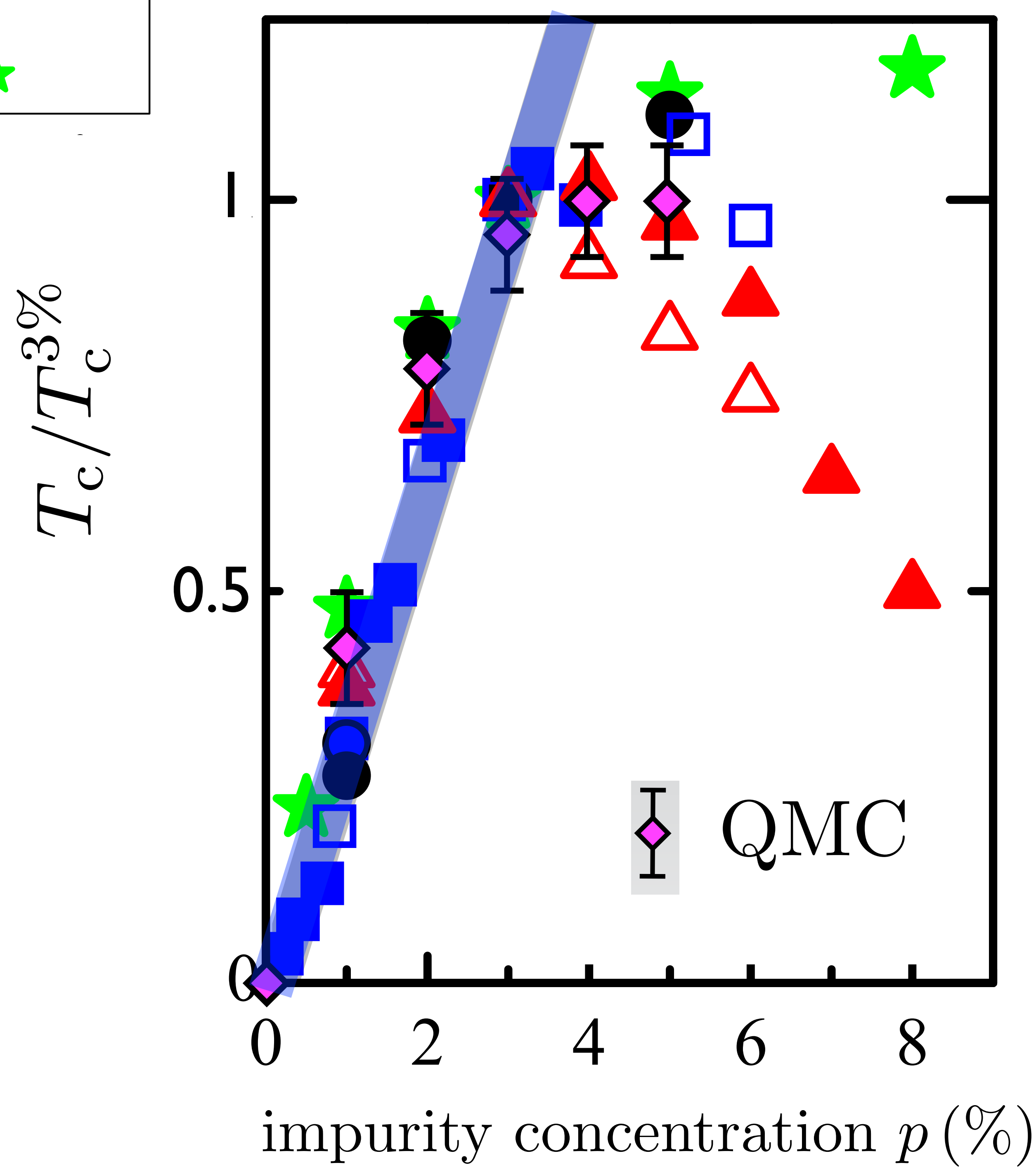

MORE PHYSICS IN THE PRESENCE OF EXTERNAL FIELD?

Spin-gap + disorder + external magnetic field

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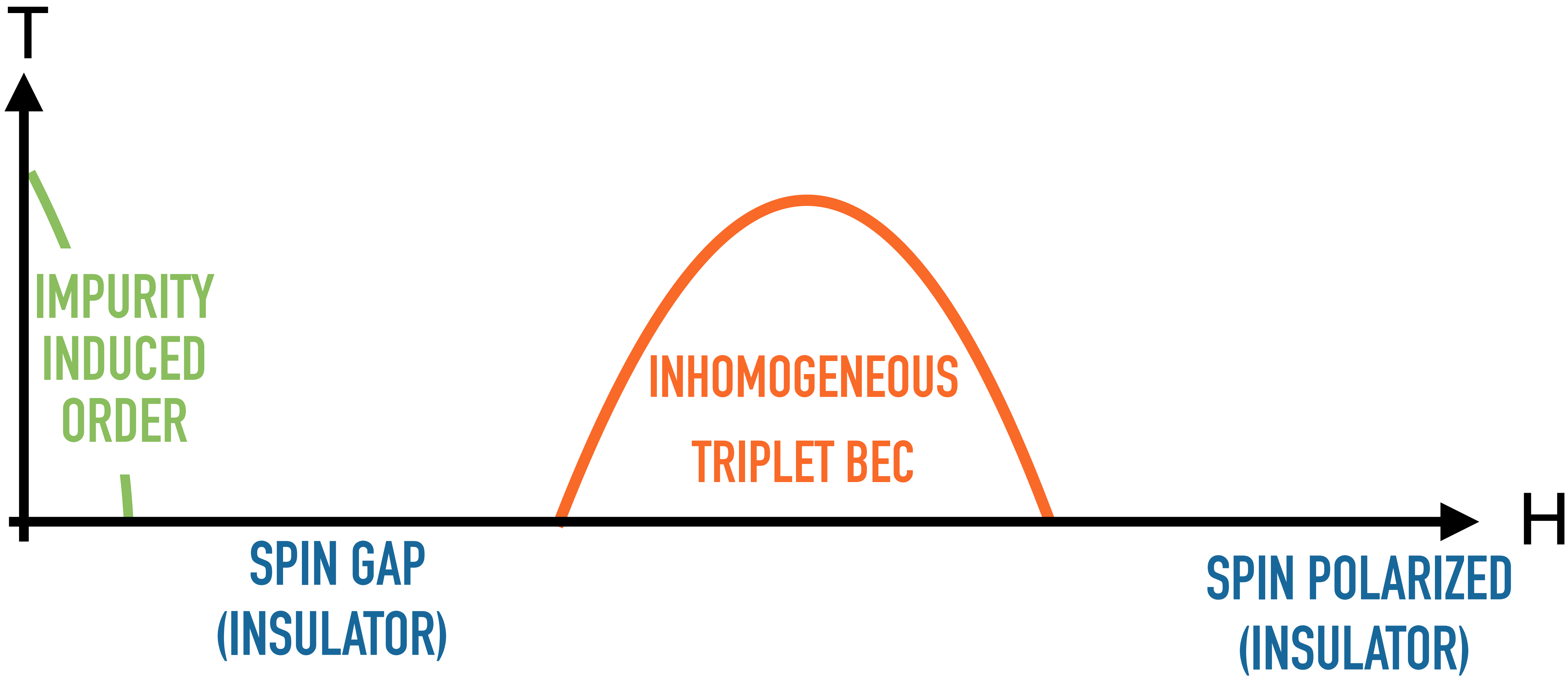


MORE PHYSICS IN THE PRESENCE OF EXTERNAL FIELD?

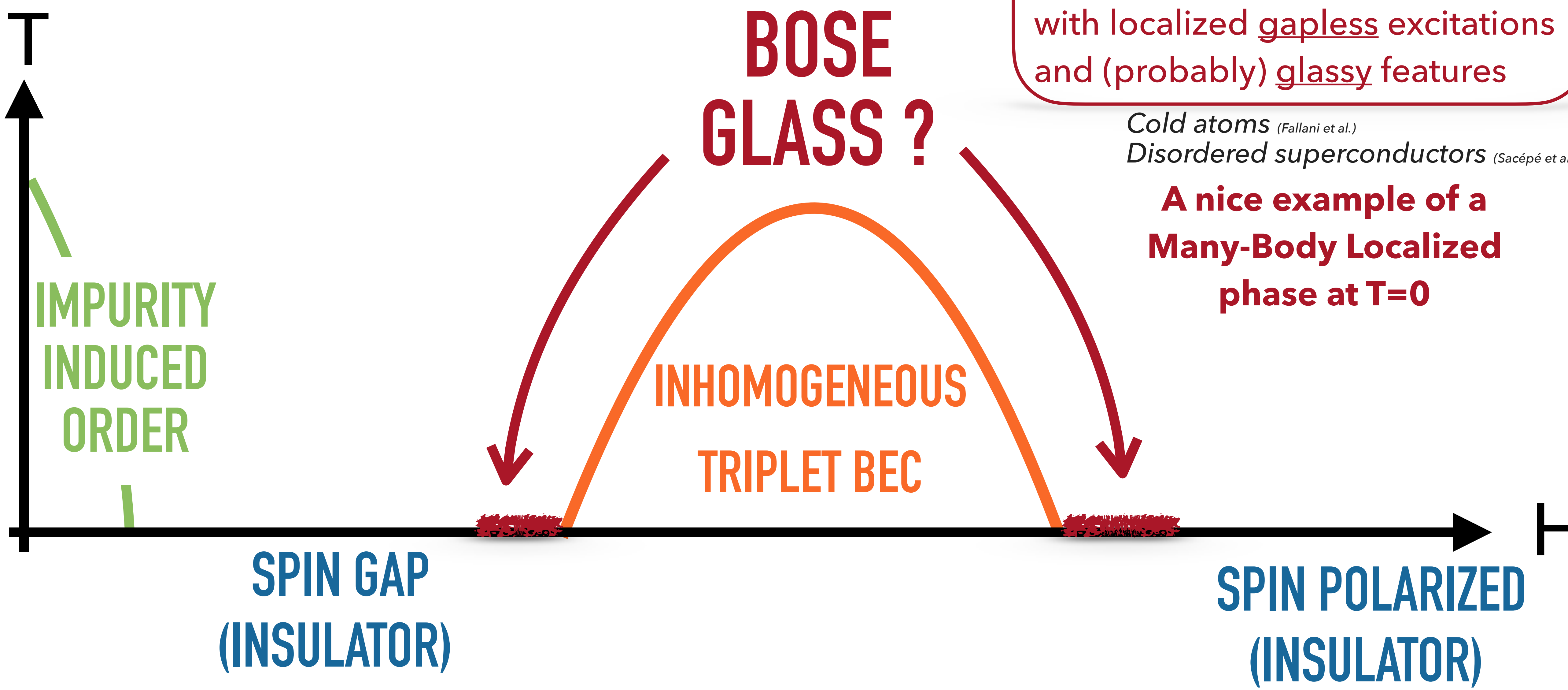
Spin-gap + disorder + external magnetic field

Giamarchi-Schulz, EPL 1987 ; Fisher *et al.* PRB 1989

A disordered insulator of bosons with localized gapless excitations and (probably) glassy features

Cold atoms (Fallani et al.)
Disordered superconductors (Sacépé et al.)

A nice example of a Many-Body Localized phase at T=0



BOSE-GLASS PHYSICS IN QUANTUM MAGNETS: ONLY A FEW EXAMPLES

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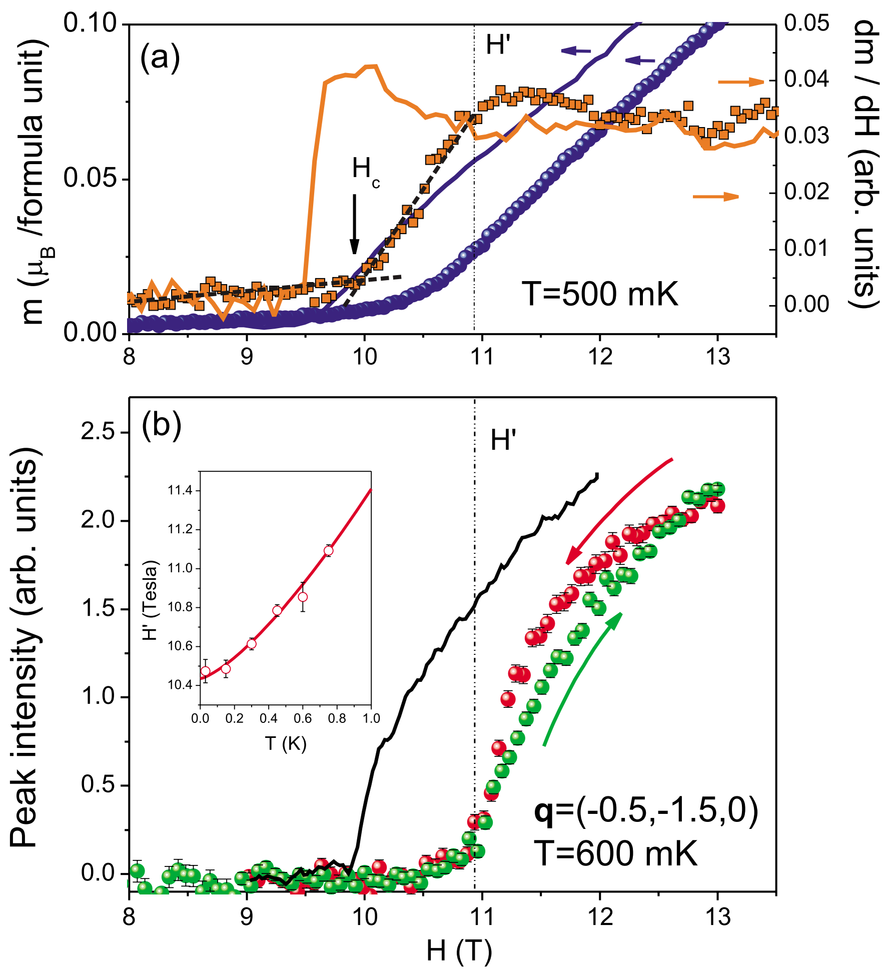
- IPACuCl3 (spin-1/2 ladders)

PHYSICAL REVIEW B **81**, 060410(R) (2010)

Evidence of a magnetic Bose glass in $(CH_3)_2CHNH_3Cu(Cl_{\{0.95\}}Br_{\{0.05\}})_3$ from neutron diffraction

Tao Hong,[1] A. Zheludev,[2,3,*] H. Manaka,[4] and L.-P. Regnault[5]



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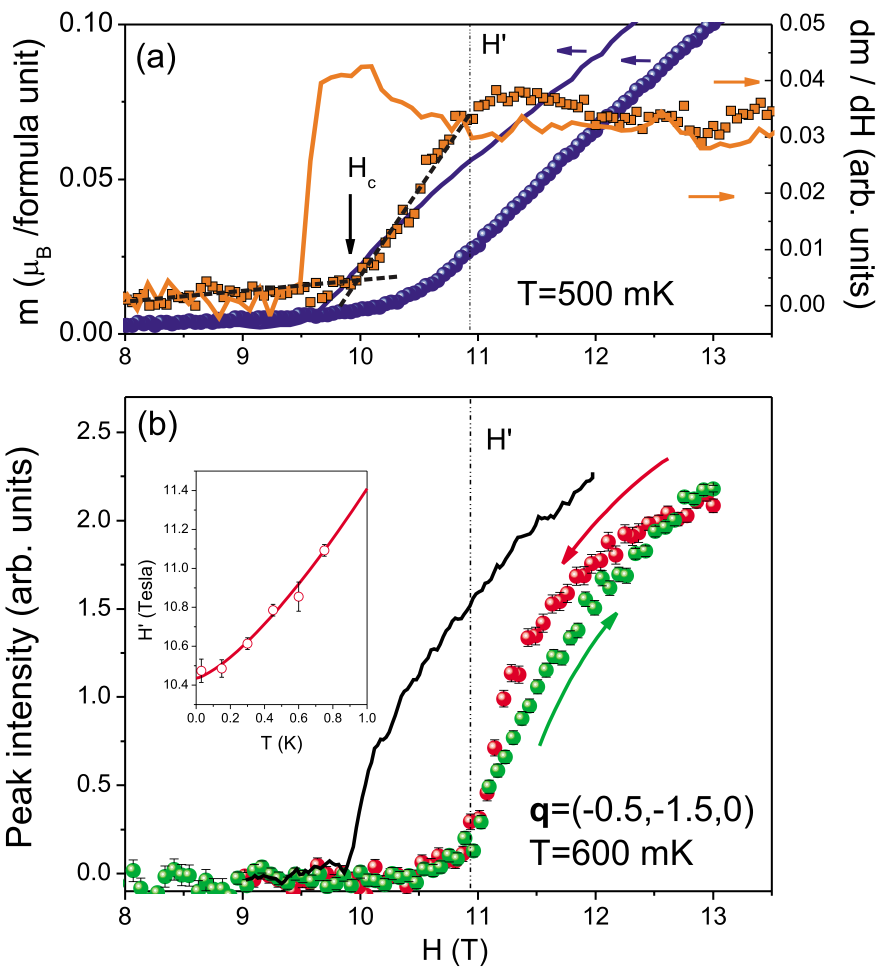


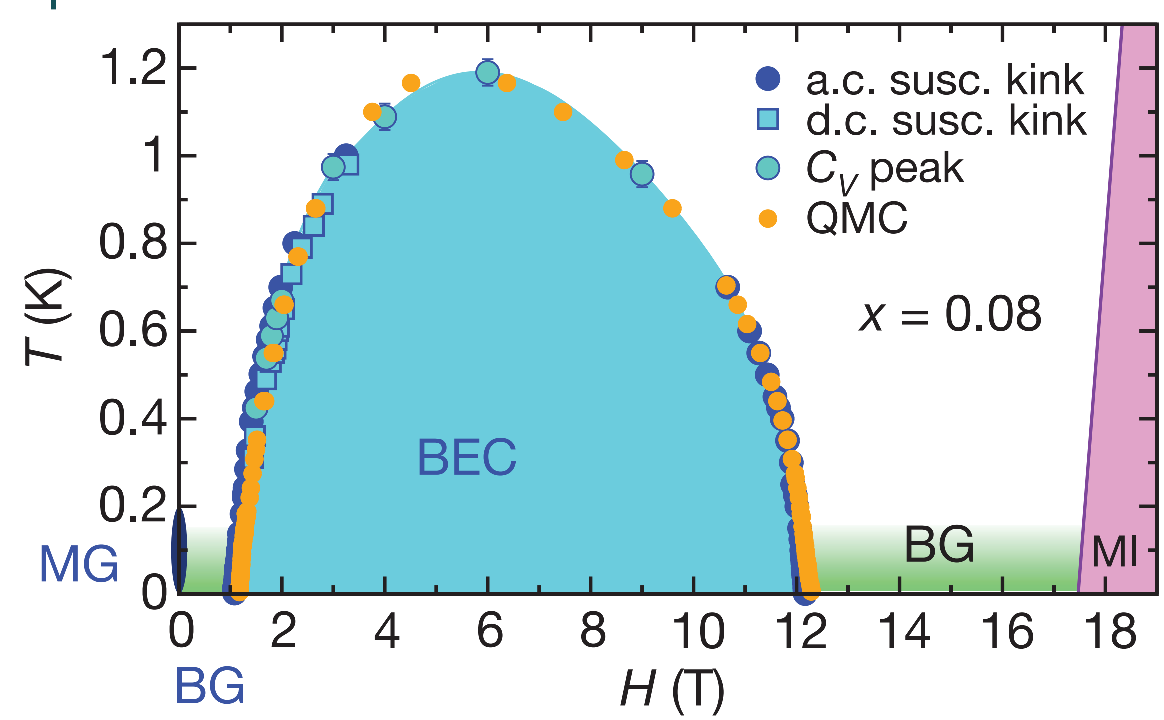
- DTN (spin-1 chains)

Nature **489**, 379 (2012)

doi:10.1038/nature11406

Bose glass and Mott glass of quasiparticles in a doped quantum magnet

Rong Yu[1], Liang Yin[2], Neil S. Sullivan[2], J. S. Xia[2], Chao Huan[2], Armando Paduan-Filho[3], Nei F. Oliveira Jr[3], Stephan Haas[4], Alexander Steppke[5], Corneliu F. Miclea[6,7], Franziska Weickert[6], Roman Movshovich[6], Eun-Deok Mun[6], Brian L. Scott[6], Vivien S. Zapf[6] & Tommaso Roscilde[8]



DTN: A PERFECT PLAYGROUND FOR FIELD-INDUCED BEC PHYSICS

$Ni(Cl)_2 - 4SC(NH_2)_2$ dichlorotetrakis-thiourea nickel

spin S=1 carrier

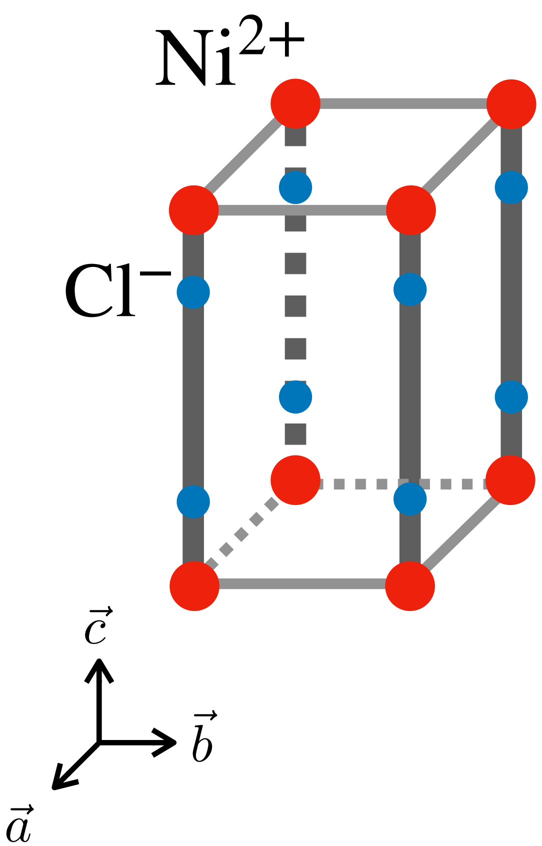
- 3D (weakly) coupled S=1chains

$$\hat{\mathcal{H}} = \underbrace{\sum_{\vec{r}} [g]\mu_B \vec{S}_{\vec{r}} \cdot \vec{H}_0 + D S^{z2}_{\vec{r}}}_{\hat{\mathcal{H}}_{loc}} + \underbrace{\sum_{\vec{r}} J_c \vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{c}}}_{\hat{\mathcal{H}}_{1D}} + \underbrace{\sum_{\vec{r}} \sum_{\vec{v}=\vec{a},\vec{b}} J_{\vec{v}} \vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{v}}}_{\hat{\mathcal{H}}_{3D}}$$

$D = 8.9$ K

$J_c = 2.2$ K

$J_{ab} = J_{3D} = 0.18$ K



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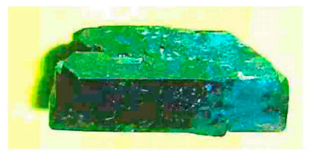

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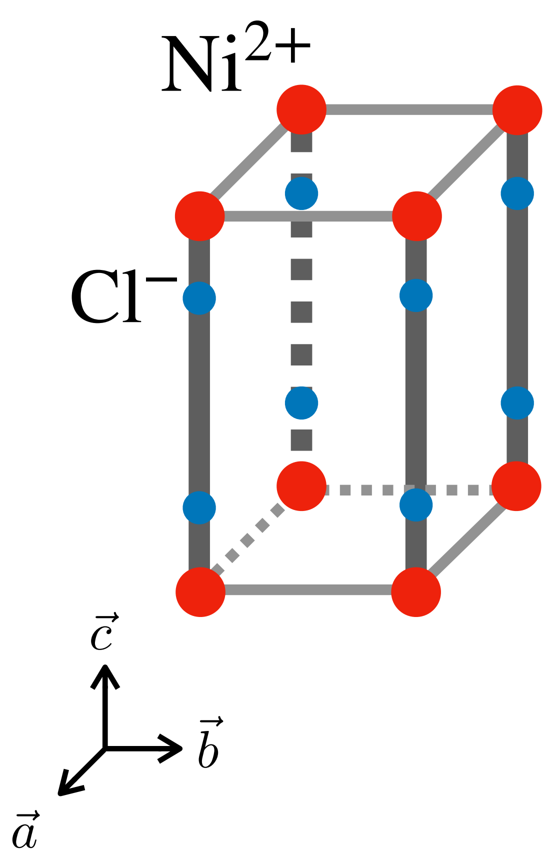

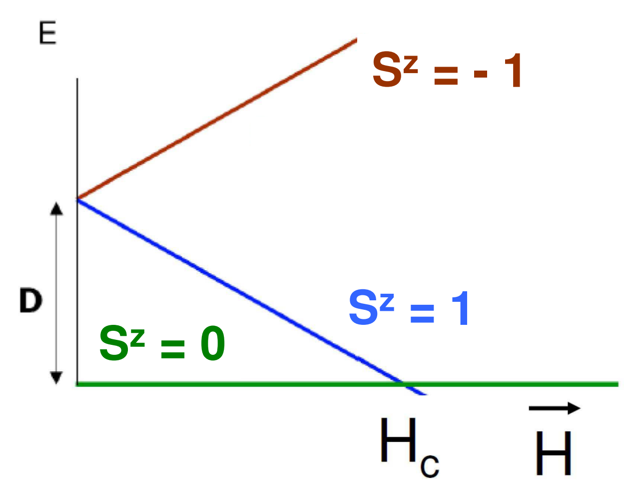

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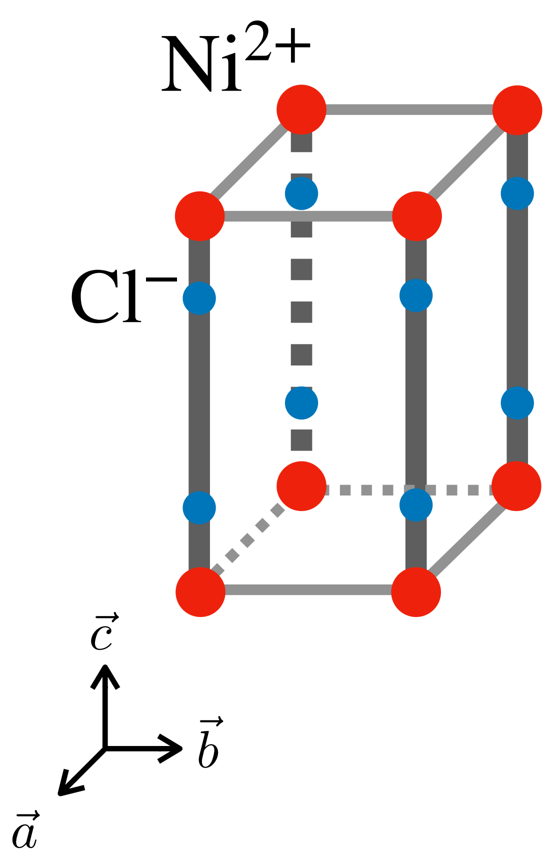

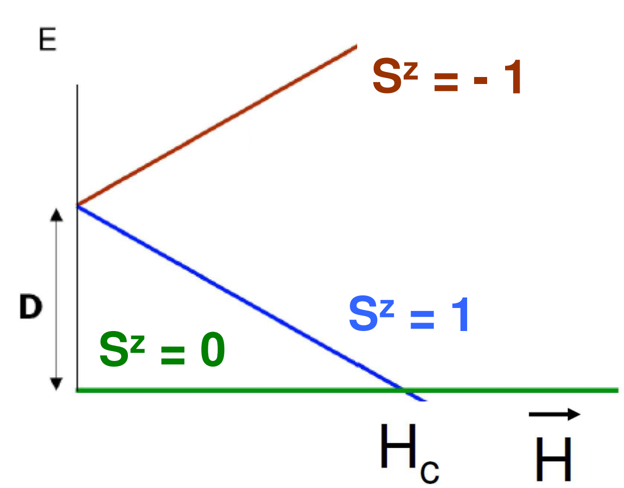

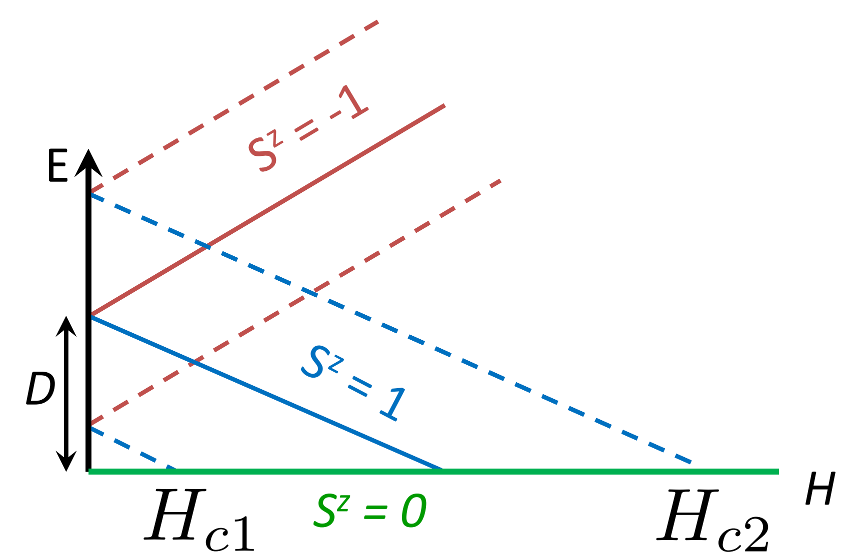

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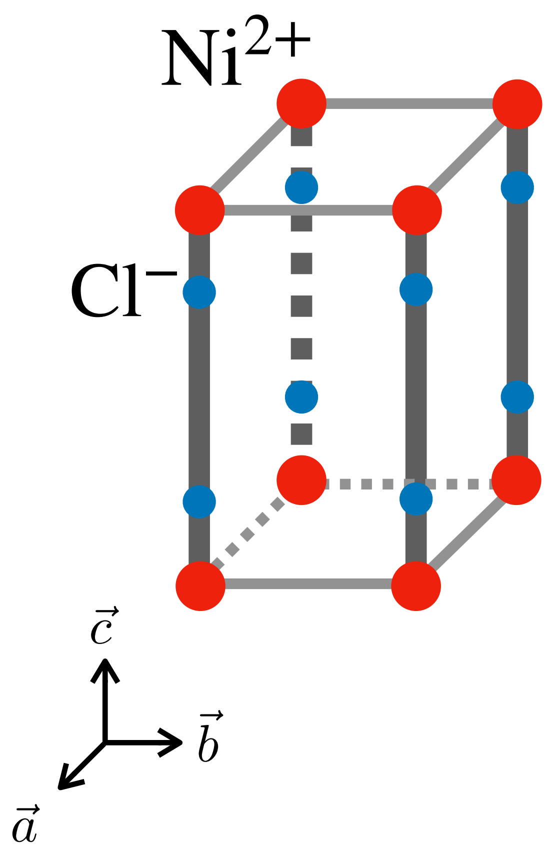
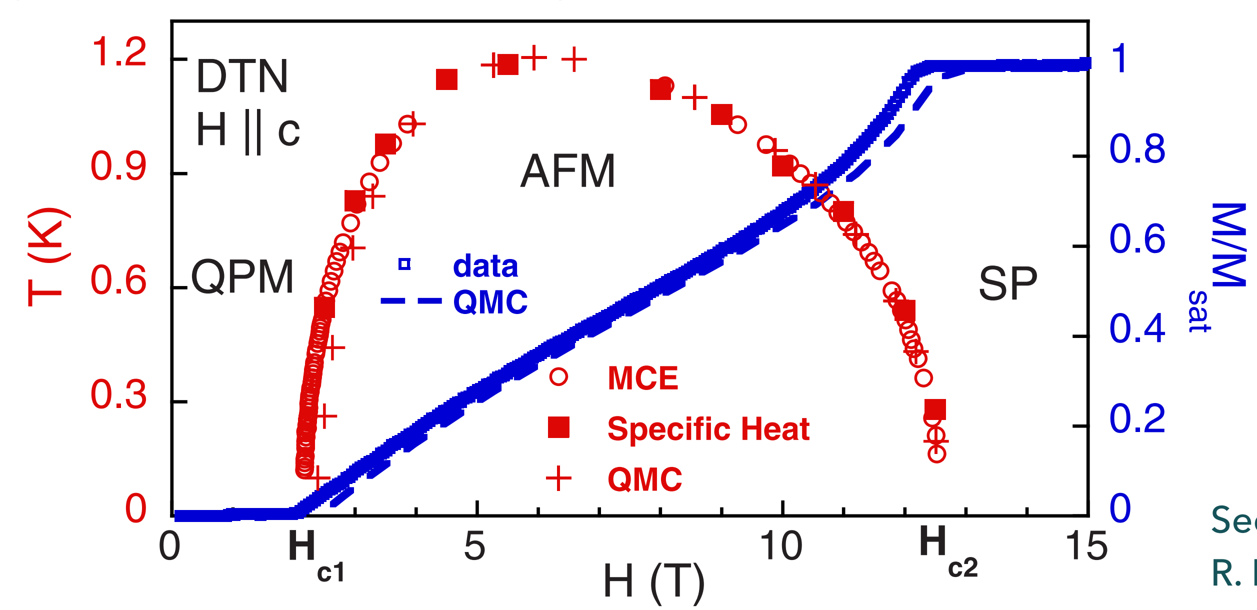


Bose-Einstein Condensation of $S = 1$ Nickel Spin Degrees of Freedom in $\mathrm{NiCl_2}$-$\mathrm{4SC(NH_2)_2}$

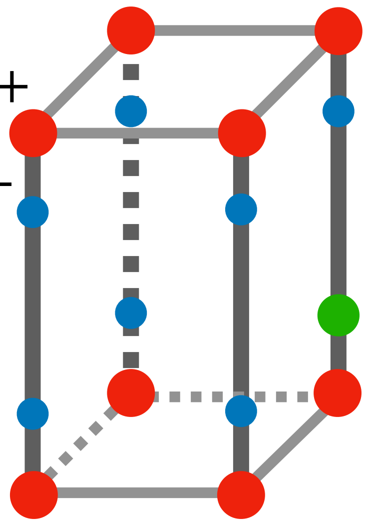
V. S. Zapf, D. Zocco, B. R. Hansen, M. Jaime, N. Harrison, C. D. Batista, M. Kenzelmann, C. Niedermayer, A. Lacerda, and A. Paduan-Filho

Phys. Rev. Lett. **96**, 077204 – Published 23 February 2006



See also NMR in
R. Blinder *at al.* PRB(R) 2017

THE DISORDERED VERSION: DTNX

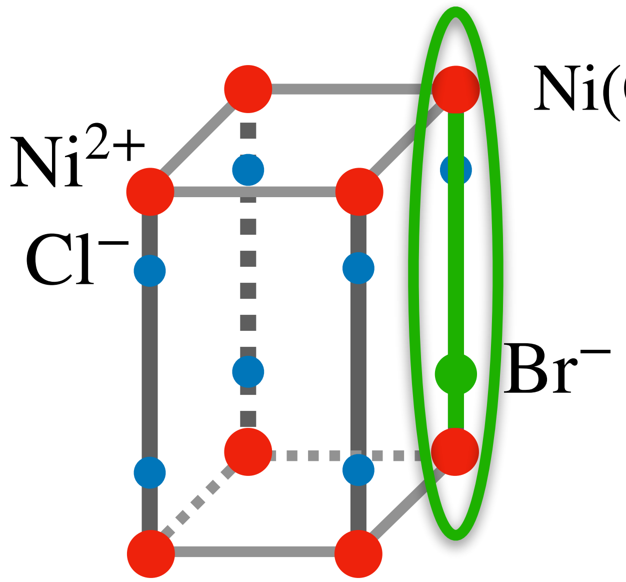


$Ni(Cl_{1-x}Br_x)_2 - 4SC(NH_2)_2$

Cl^- randomly replaced by Br^-

$$\hat{\mathcal{H}} = \underbrace{\sum_{\vec{r}} [g]\mu_B \vec{S}_{\vec{r}} \cdot \vec{H}_0 + D S_{\vec{r}}^{z\,2}}_{\hat{\mathcal{H}}_{loc}} + \underbrace{\sum_{\vec{r}} J_c\, \vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{c}}}_{\hat{\mathcal{H}}_{1D}} + \underbrace{\sum_{\vec{r}} \sum_{\vec{v}=\vec{a},\vec{b}} J_{\vec{v}}\, \vec{S}_{\vec{r}} \cdot \vec{S}_{\vec{r}+\vec{v}}}_{\hat{\mathcal{H}}_{3D}}$$

THE DISORDERED VERSION: DTNX



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Doping does **not introduce non-magnetic impurities**

It is reasonable to assume that it only **modifies the couplings** in the close vicinity of a Br

MICROSCOPIC PARAMETERS FROM NMR EXPERIMENTS

A. Orlova *et al.* PRL 2017
M. Dupont *et al.* PRB 2017

$\mathrm{Ni(Cl_{0.96}Br_{0.04})_2 - 4SC(NH_2)_2}$

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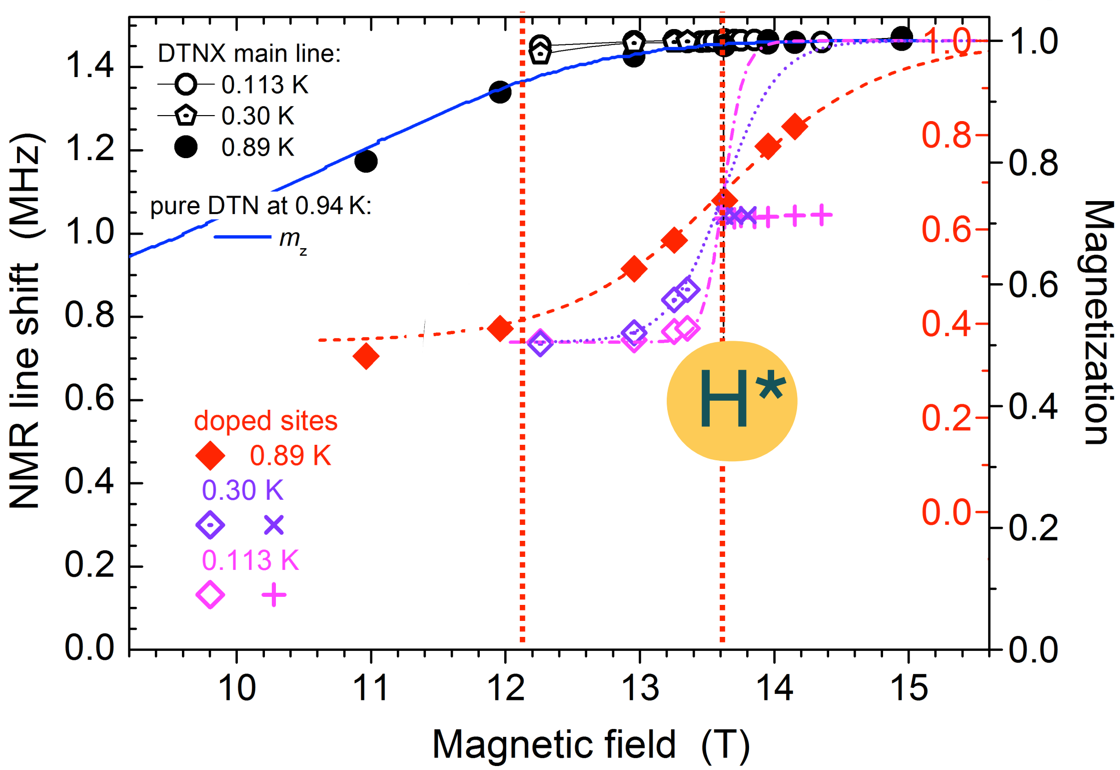
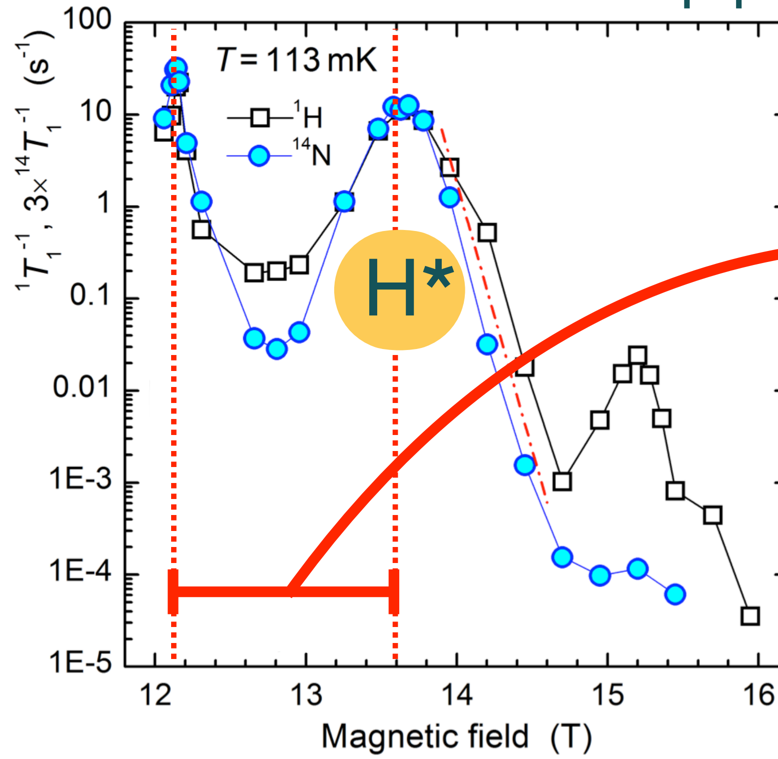
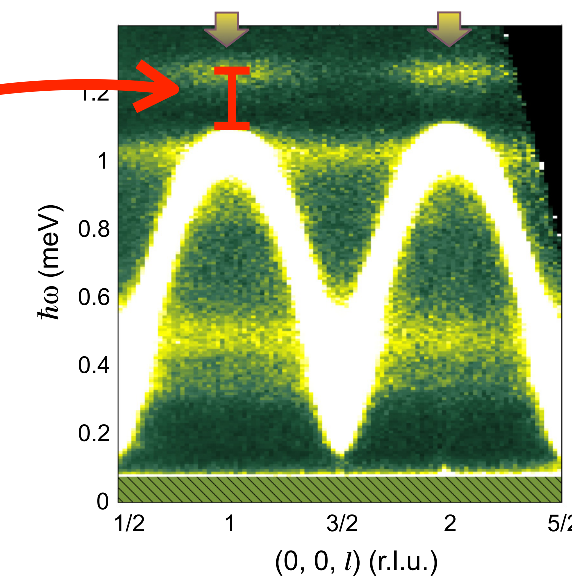


NMR relaxation: sharp peak at H*



Localized state also seen in neutrons



K. Povarov *et al.* PRB 2015

Magnetization of impurity site $m_z \approx 0.365$

Localized state above H_{c2} at $H^* \approx 13.6$T

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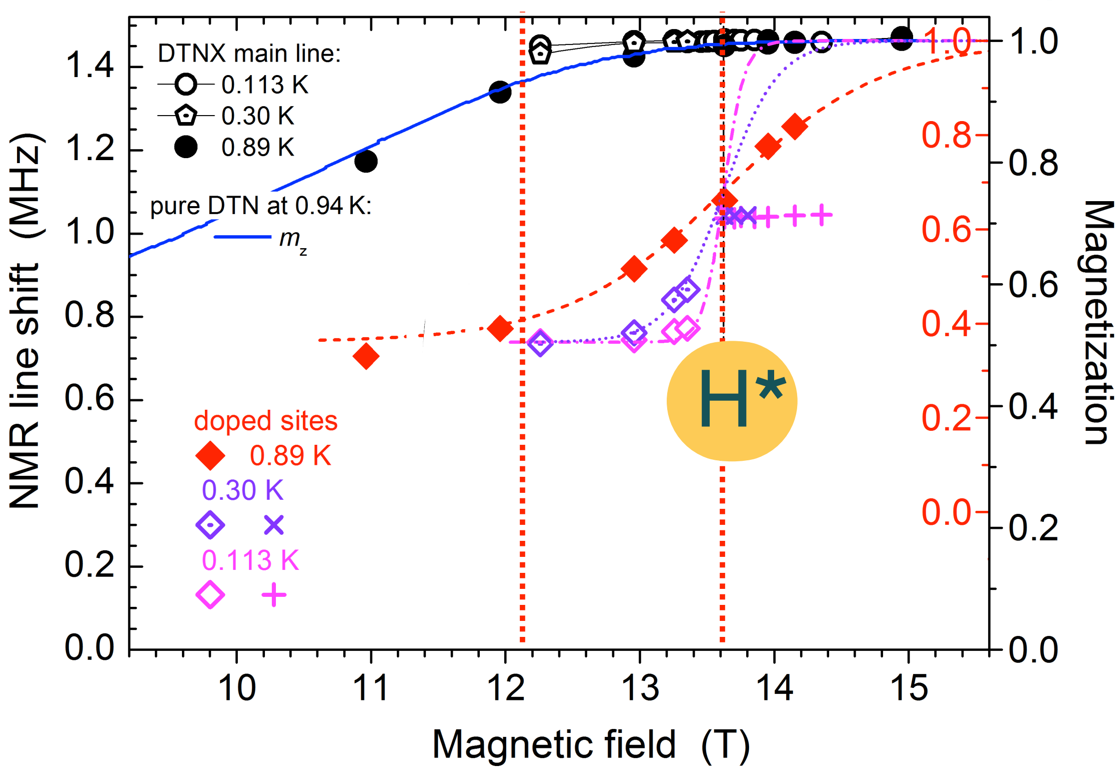

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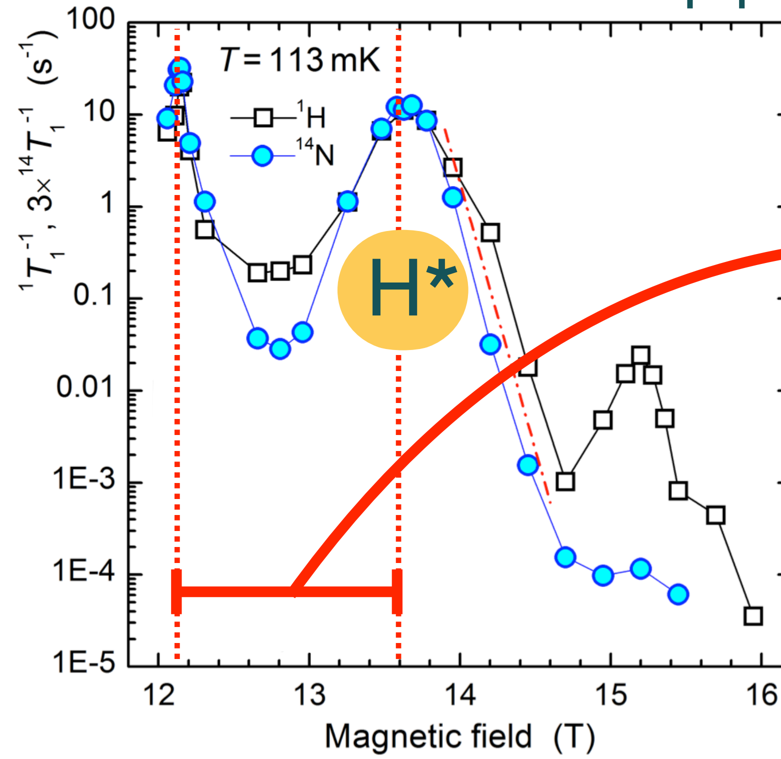

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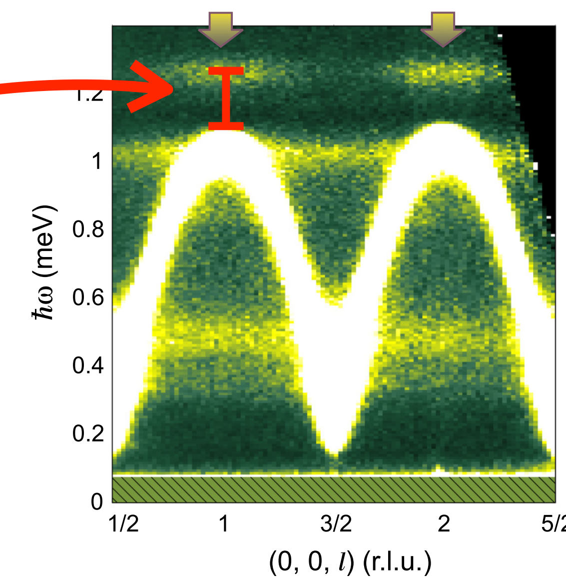

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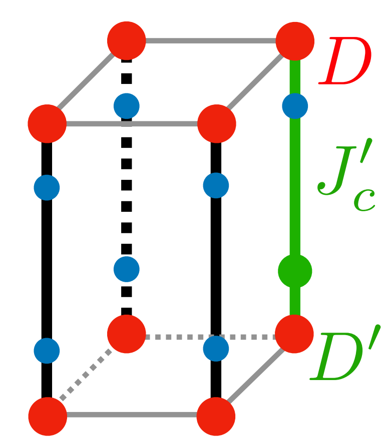
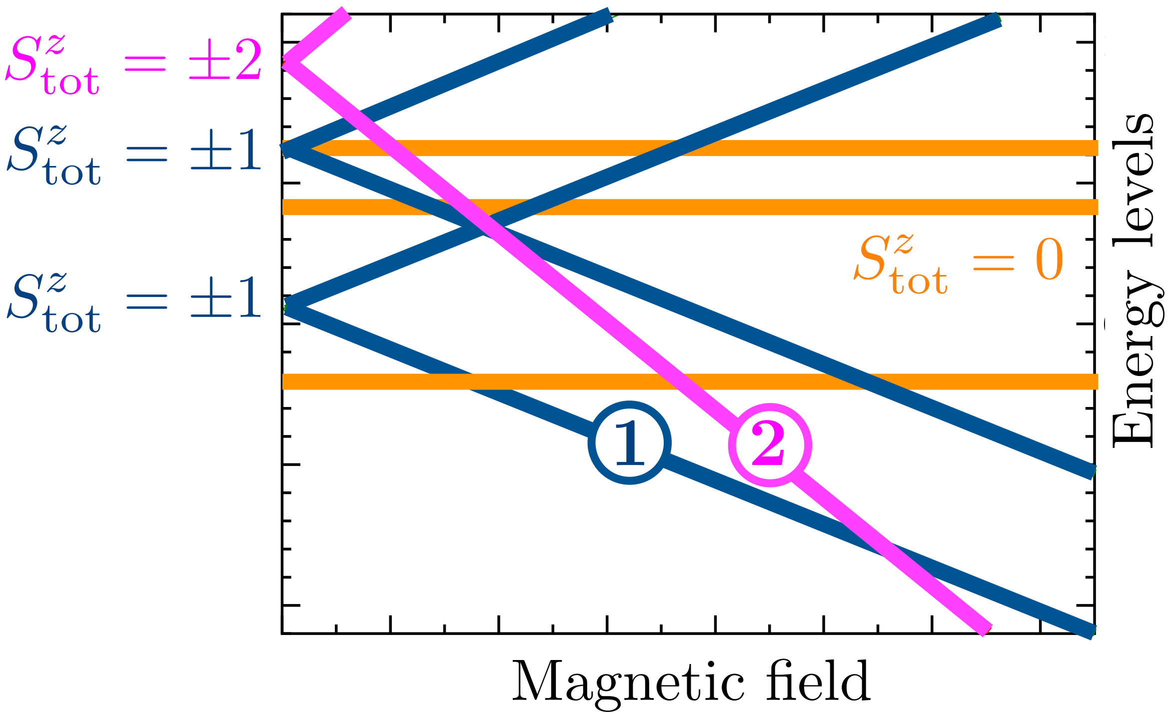
➡ Theoretical modeling

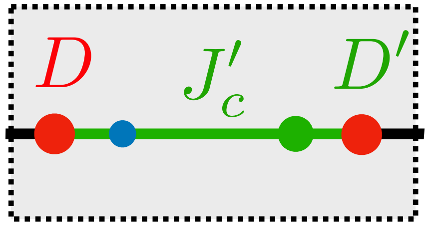


THEORETICAL MODELLING

M. Dupont *et al.* PRB 2017

A single perturbed dimer

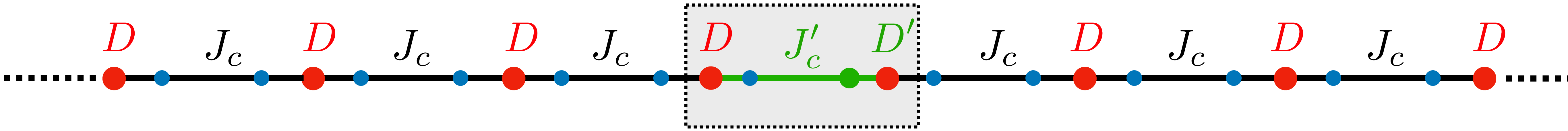




$$\mathcal{H}_0 = D S^z_1 + D' S^z_2 + J'_c \vec{S}_1 \cdot \vec{S}_2 - g\mu_B H \left(S^z_1 + S^z_2\right)$$

THEORETICAL MODELLING

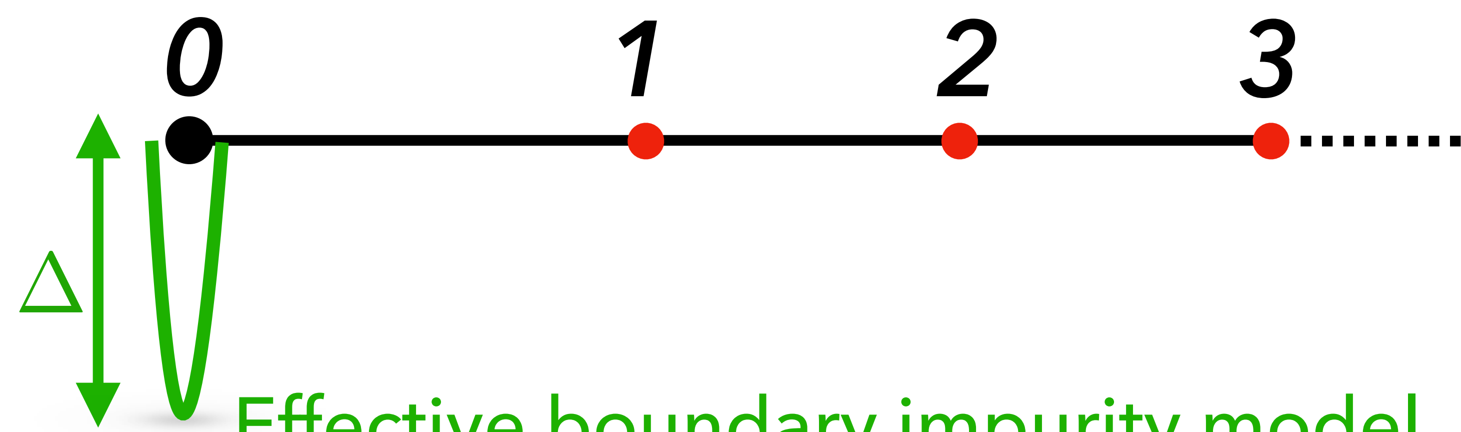
M. Dupont *et al.* PRB 2017



Include 1d dynamics

=> $S^z = 1$: localized bound state

$$\xi = \frac{1}{2\ln(\Delta/J_c)}$$



Effective boundary impurity model

$$\mathcal{H}_{\text{eff}} = -\Delta|0\rangle\langle 0| + J_c \sum_{j'=0}^{\infty} |j'\rangle\langle j'+1| + |j'+1\rangle\langle j'|$$

$$\Delta = J'_c - J_c + \frac{D' - D + \sqrt{(D'-D)^2 + (2J'_c)^2}}{2}$$

THEORETICAL MODELLING

M. Dupont *et al.* PRB 2017

3d dynamics

$$\xi_\perp = \frac{1}{2\,\mathrm{asinh}(\Delta/2J_\perp)}$$

$$\xi = \frac{1}{2\ln(\Delta/J_c)}$$

$$H^* = H^*_{\mathrm{dimer}} + J_c + J_c \exp\left(-\frac{1}{2\xi}\right) + 4J_\perp + 4J_\perp \exp\left(-\frac{1}{2\xi_\perp}\right)$$

THEORETICAL MODELLING

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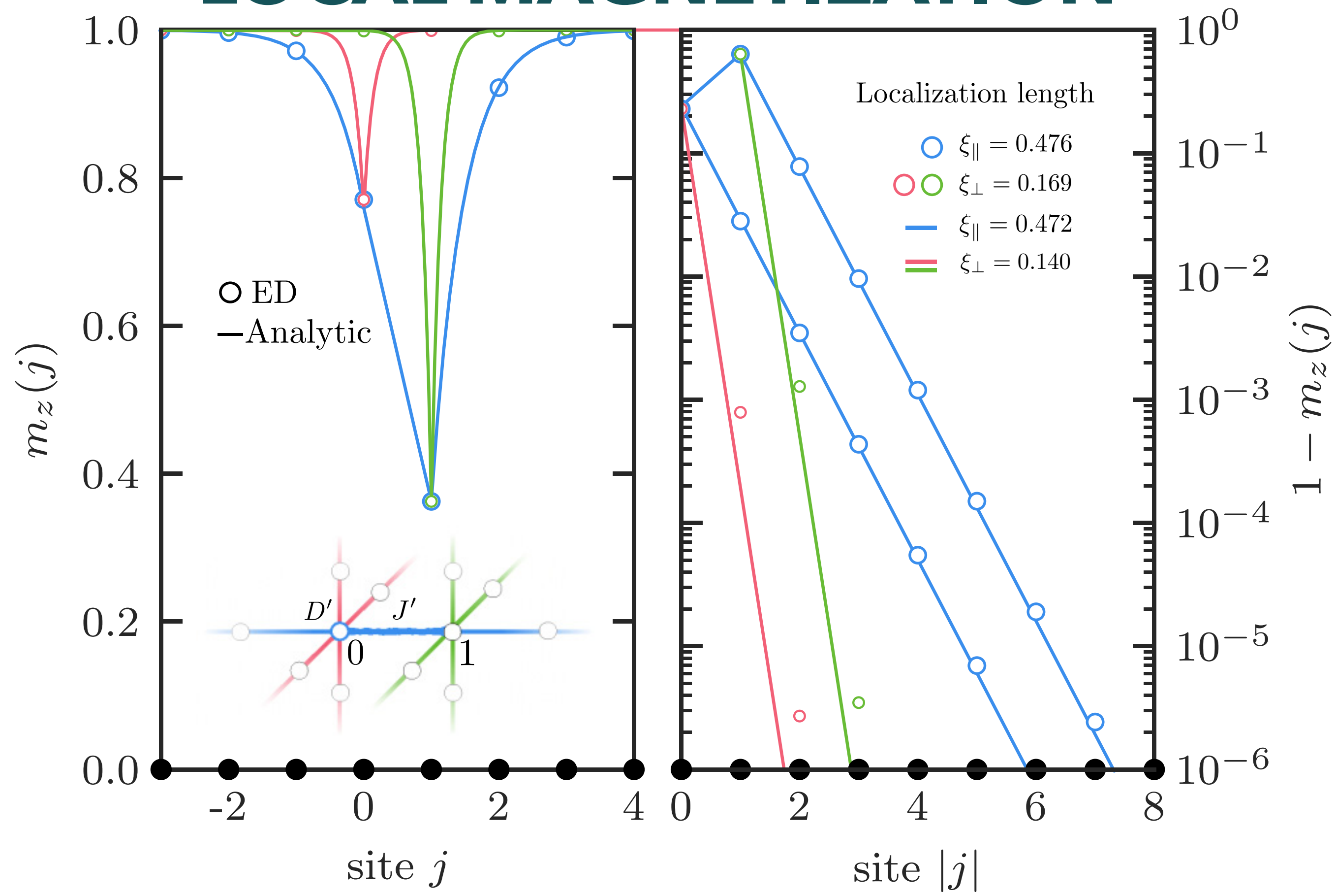
LOCAL MAGNETIZATION

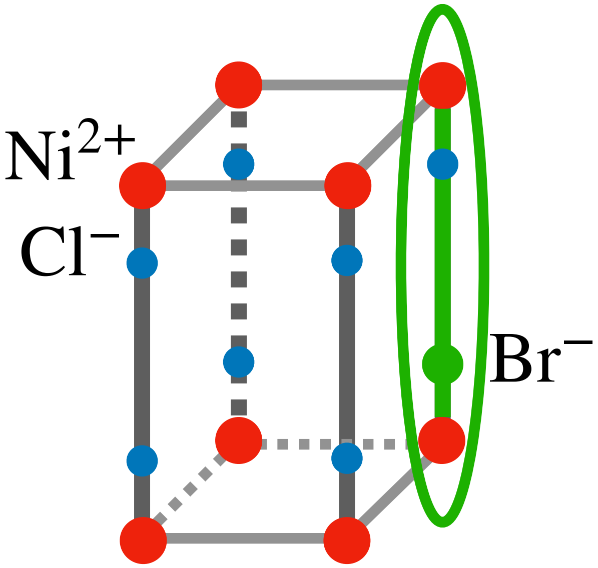
We also have analytical expressions for the local magnetisations: Nicely comparable to ED!



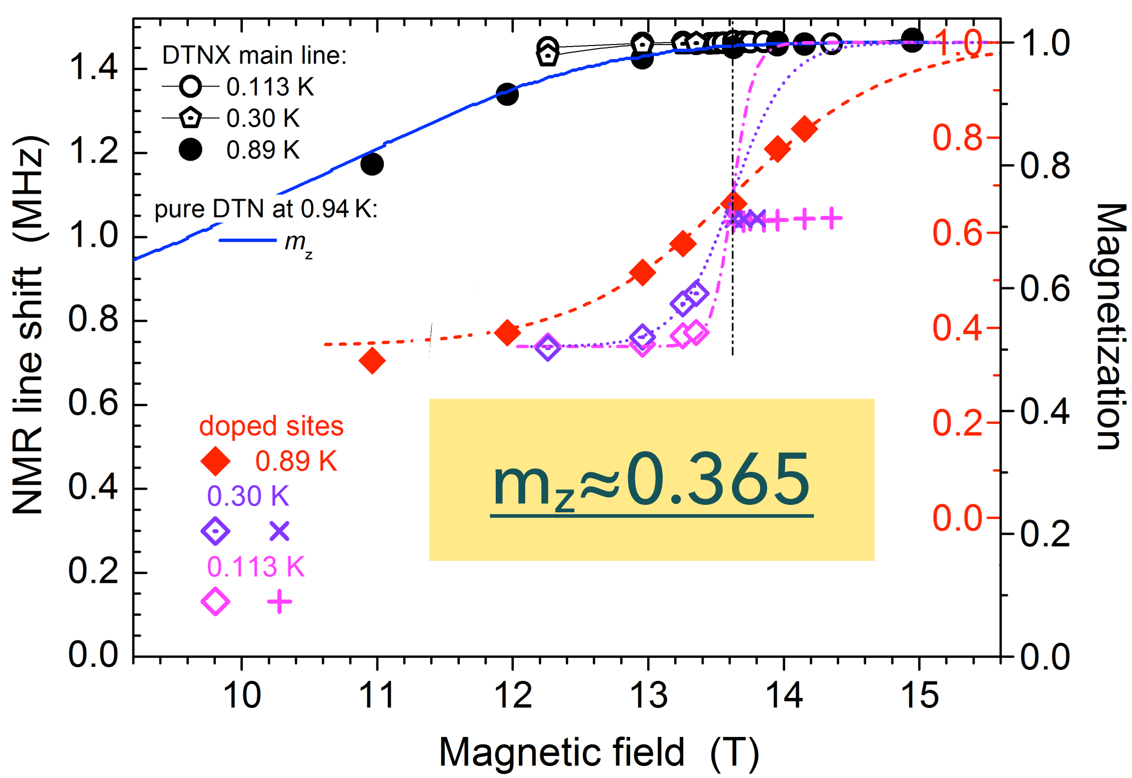
The $S^z = 1$ state is strongly localized on the perturbed dimer!

THEORETICAL MODELLING: SUMMARY

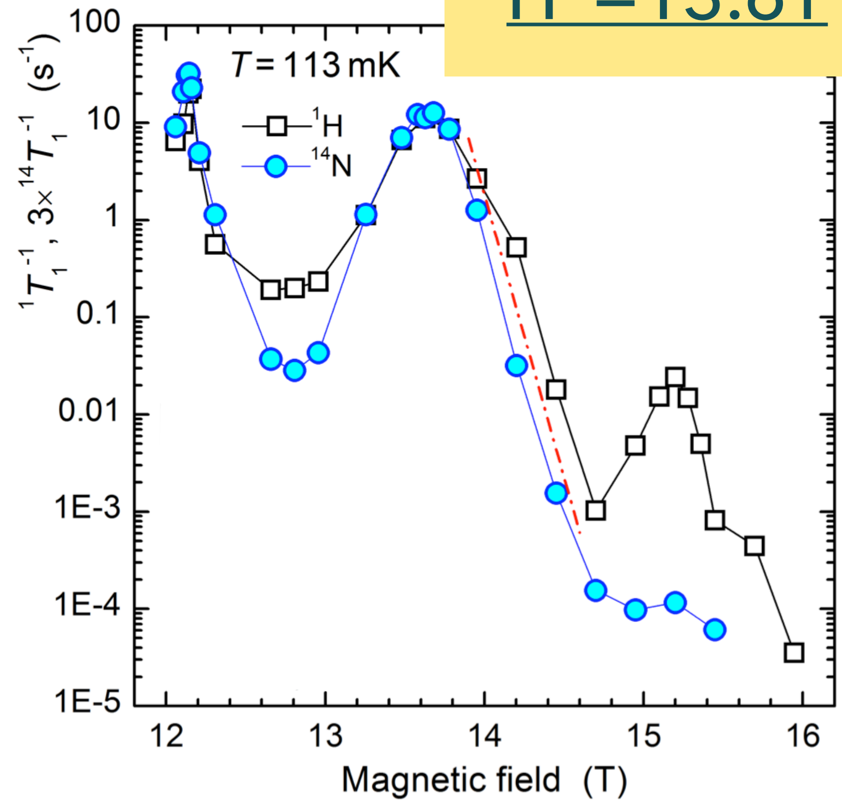
M. Dupont *et al.* PRB 2017



$$\hat{\mathcal{H}} = \underbrace{\sum_{\vec{r}}[g]\mu_B \vec{S}_{\vec{r}}\cdot\vec{H}_0 + D' S_{\vec{r}}^{z2}}_{\hat{\mathcal{H}}_{loc}} + \underbrace{\sum_{\vec{r}} J'_c\, \vec{S}_{\vec{r}}\cdot \vec{S}_{\vec{r}+\vec{c}}}_{\hat{\mathcal{H}}_{1D}} + \underbrace{\sum_{\vec{r}}\sum_{\vec{v}=\vec{a},\vec{b}} J_{\vec{v}}\, \vec{S}_{\vec{r}}\cdot\vec{S}_{\vec{r}+\vec{v}}}_{\hat{\mathcal{H}}_{3D}}$$



$m_z \approx 0.365$



H*=13.6T

$$\Rightarrow J'_c = 2.42\,J_c \approx 5.3\,\mathrm{K} \quad \text{and} \quad D' = 0.36\,D \approx 3.2\,\mathrm{K}$$

QMC STUDY OF THE MOST REALISTIC 3D S=1 DTNX MODEL

M. Dupont *et al.* PRL 2017
M. Dupont *et al.* PRB 2017

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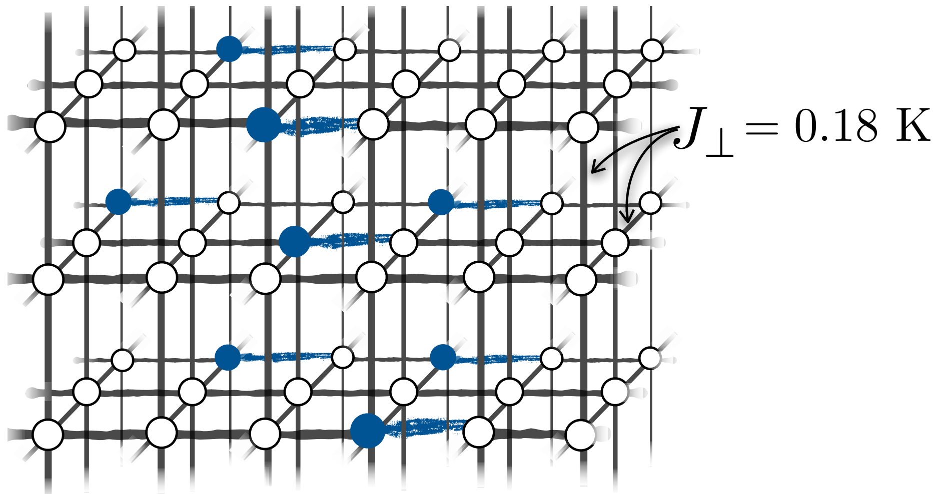
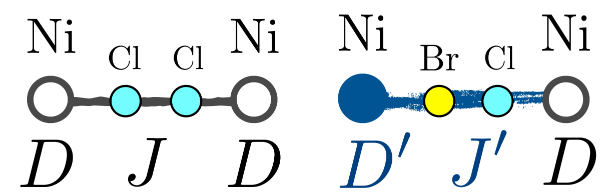
finite concentration (x) of perturbed bonds



(disorder averaging is performed)



$J = 2.2$ K

$D = 8.9$ K

$J' = 5.32$ K

$D' = 3.2$ K

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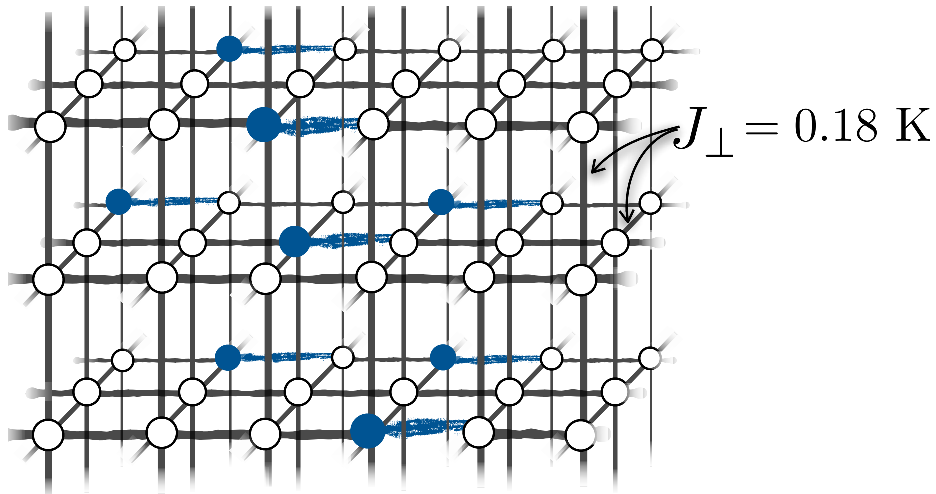

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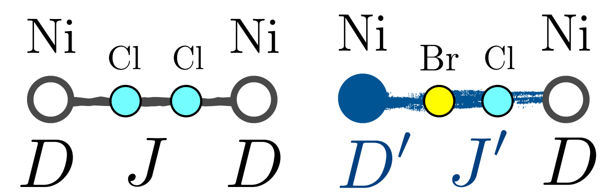

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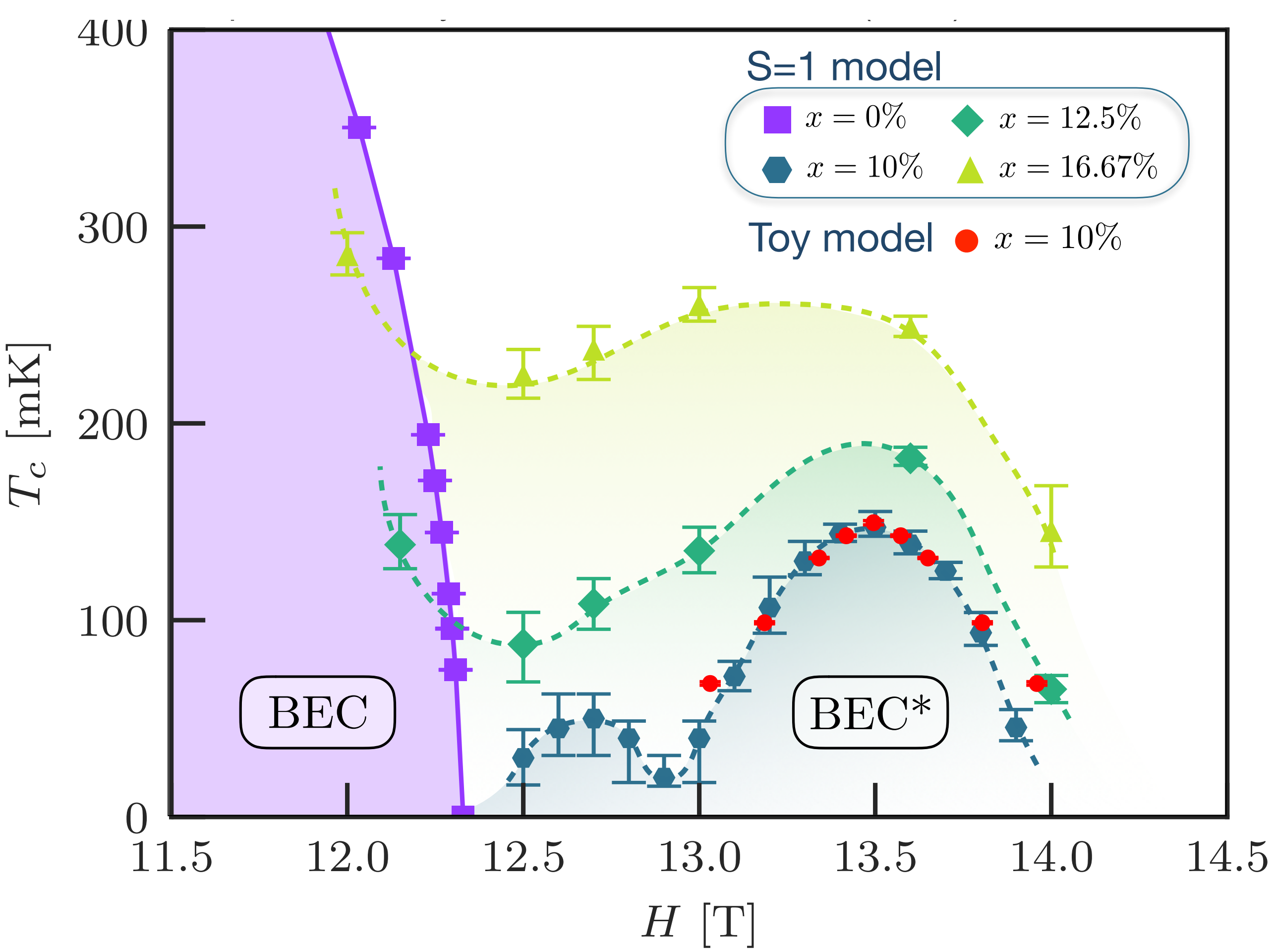
QMC phase diagram



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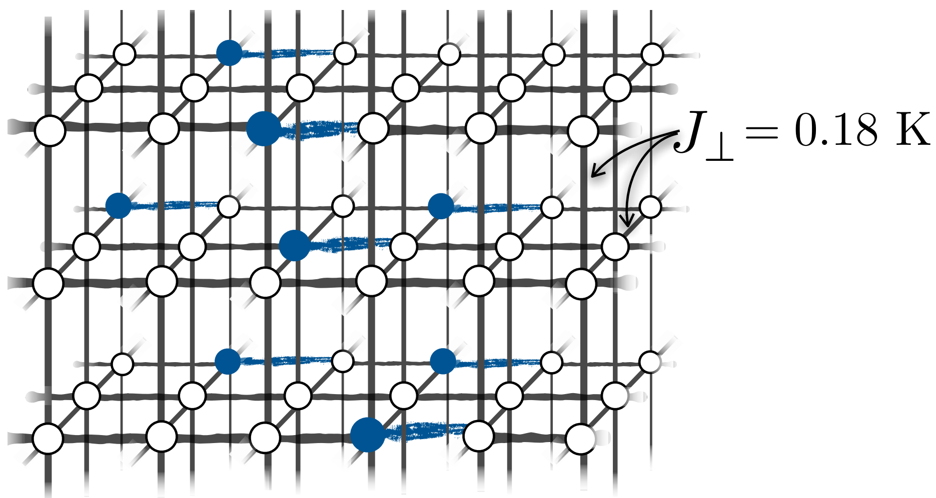

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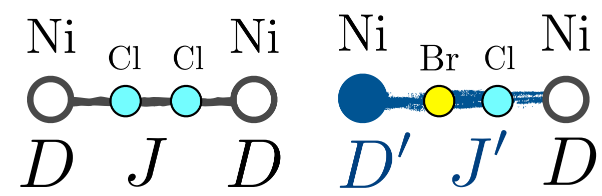

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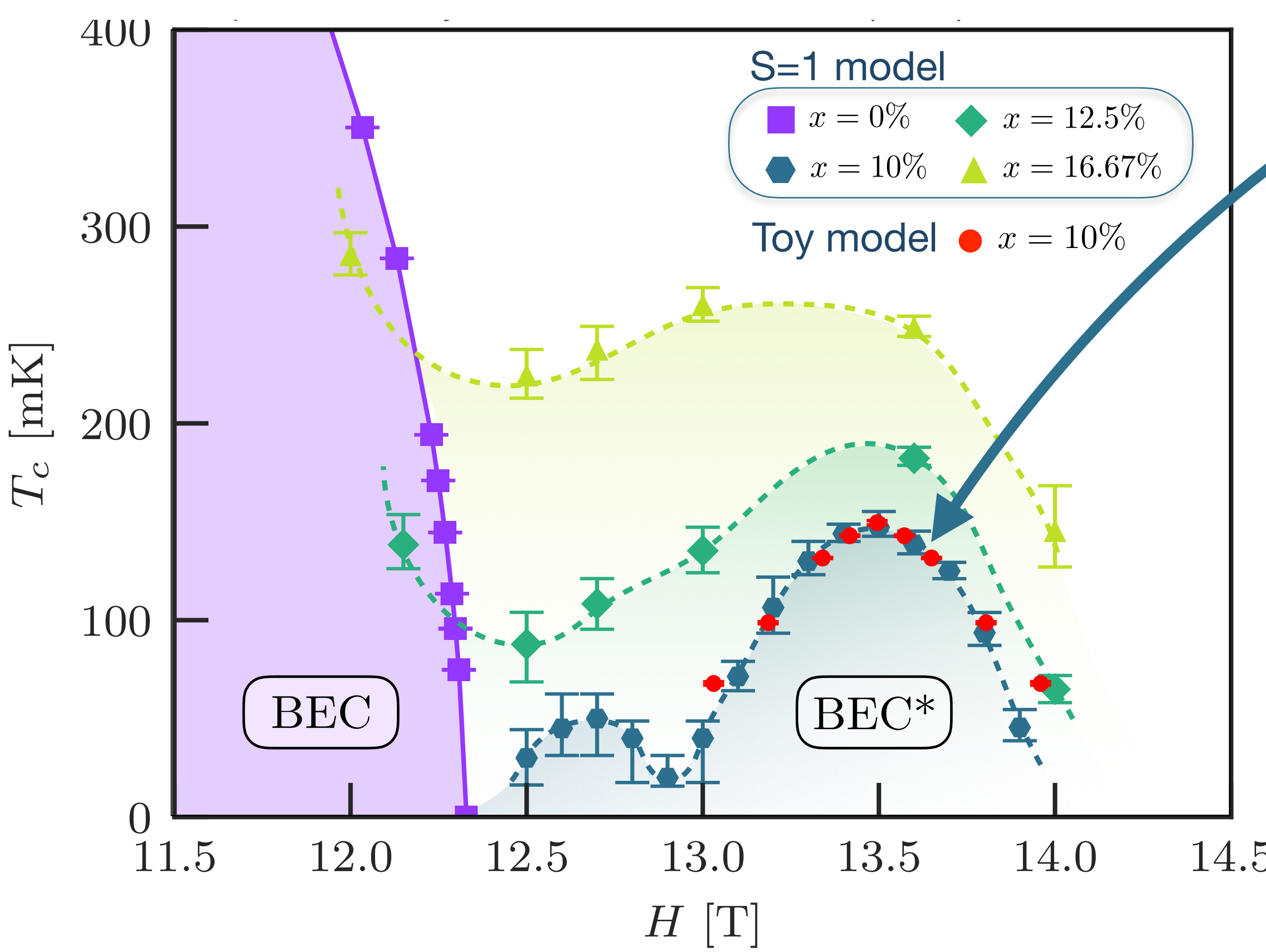
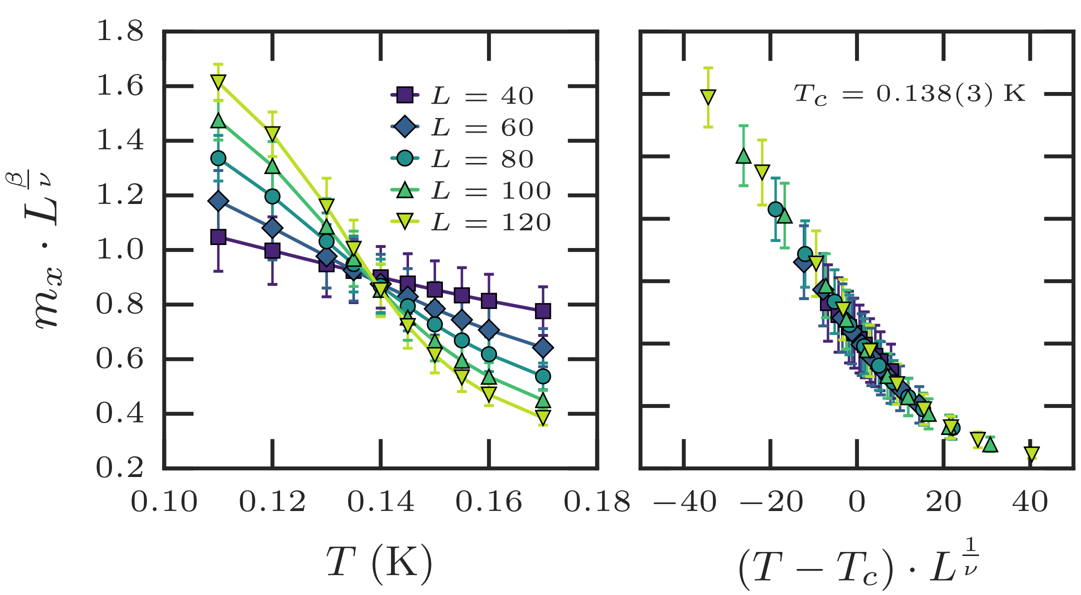


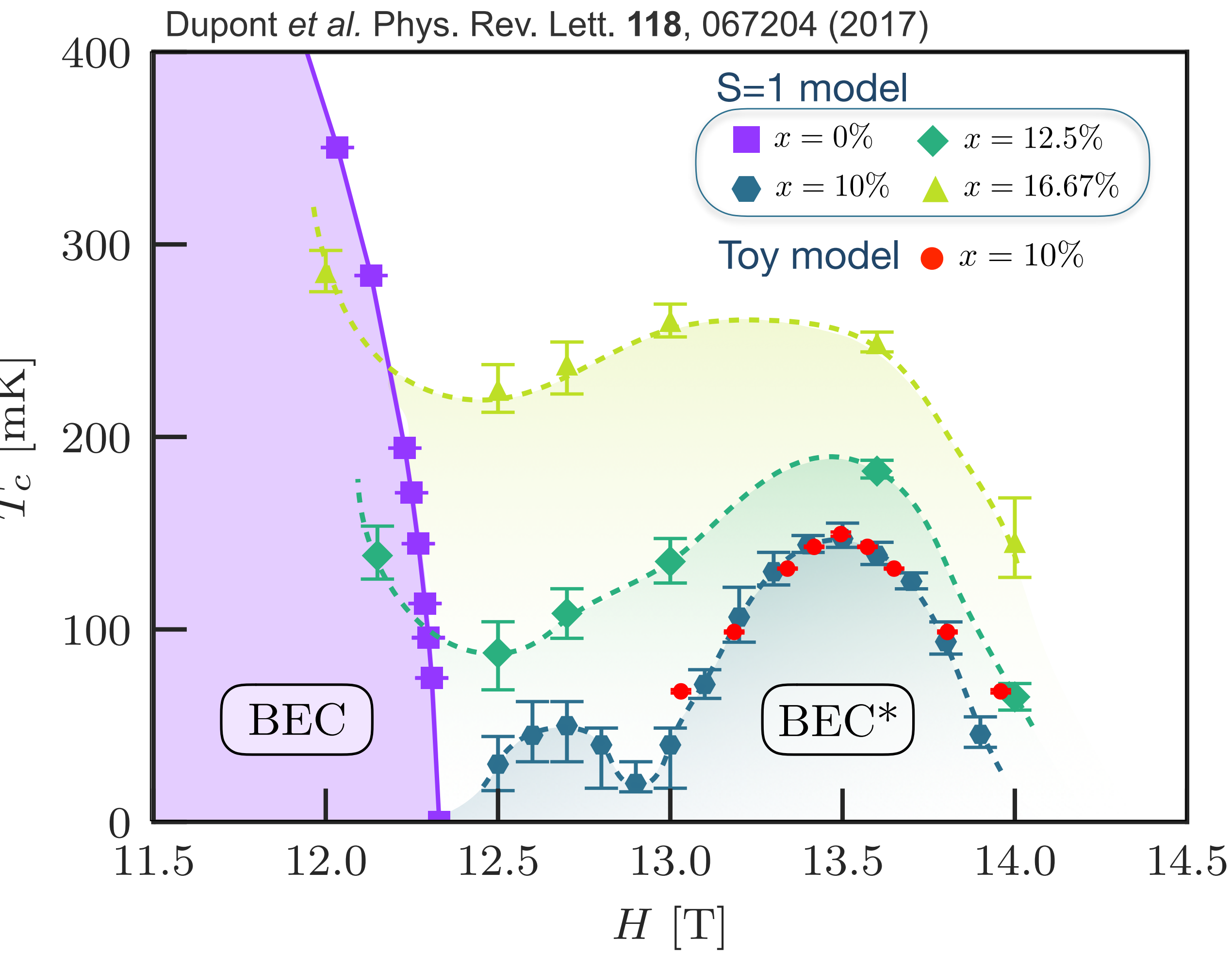
3DXY (BEC) Universality class

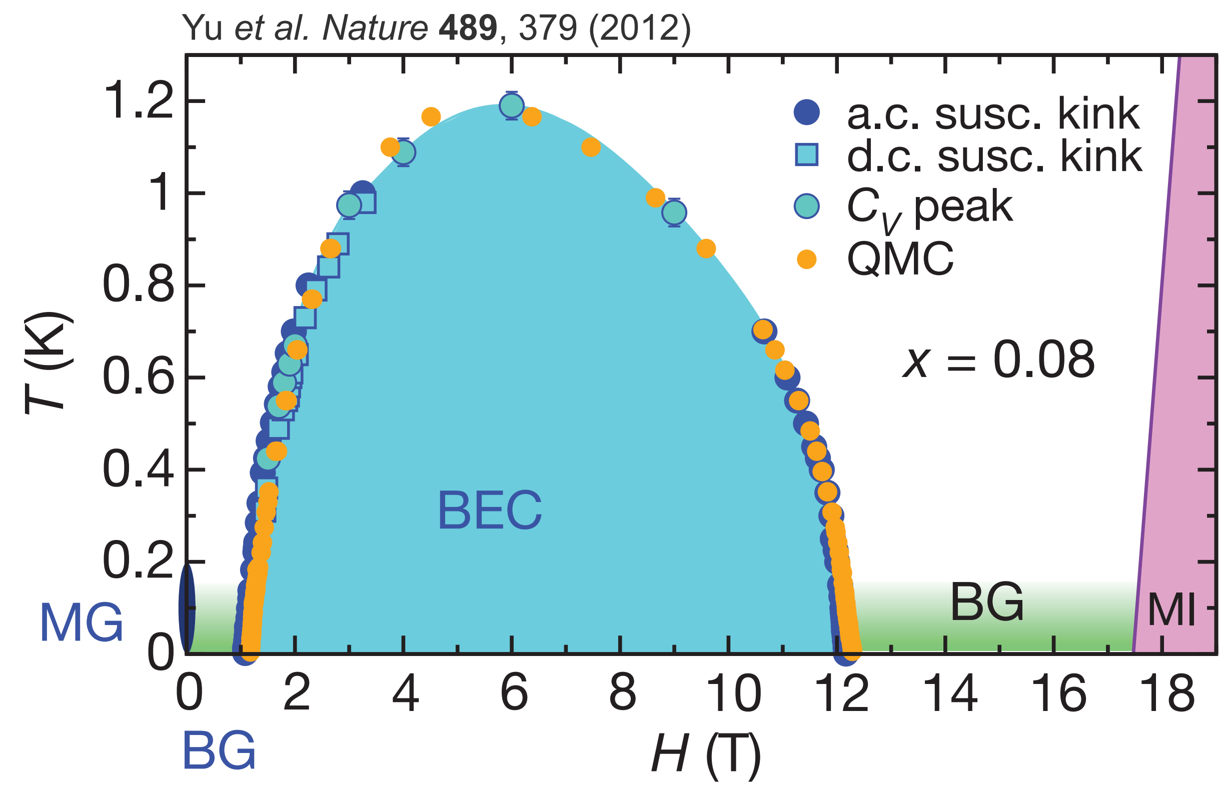
$$m^{\mathrm{AF}}(L) = L^{-\beta/\nu}\mathcal{G}_{m^{\mathrm{AF}}}\left[L^{1/\nu}(T - T_c)\right]$$

$\nu = 0.6717$ and $\beta = 0.3486$



THEORY VS EXPERIMENTS





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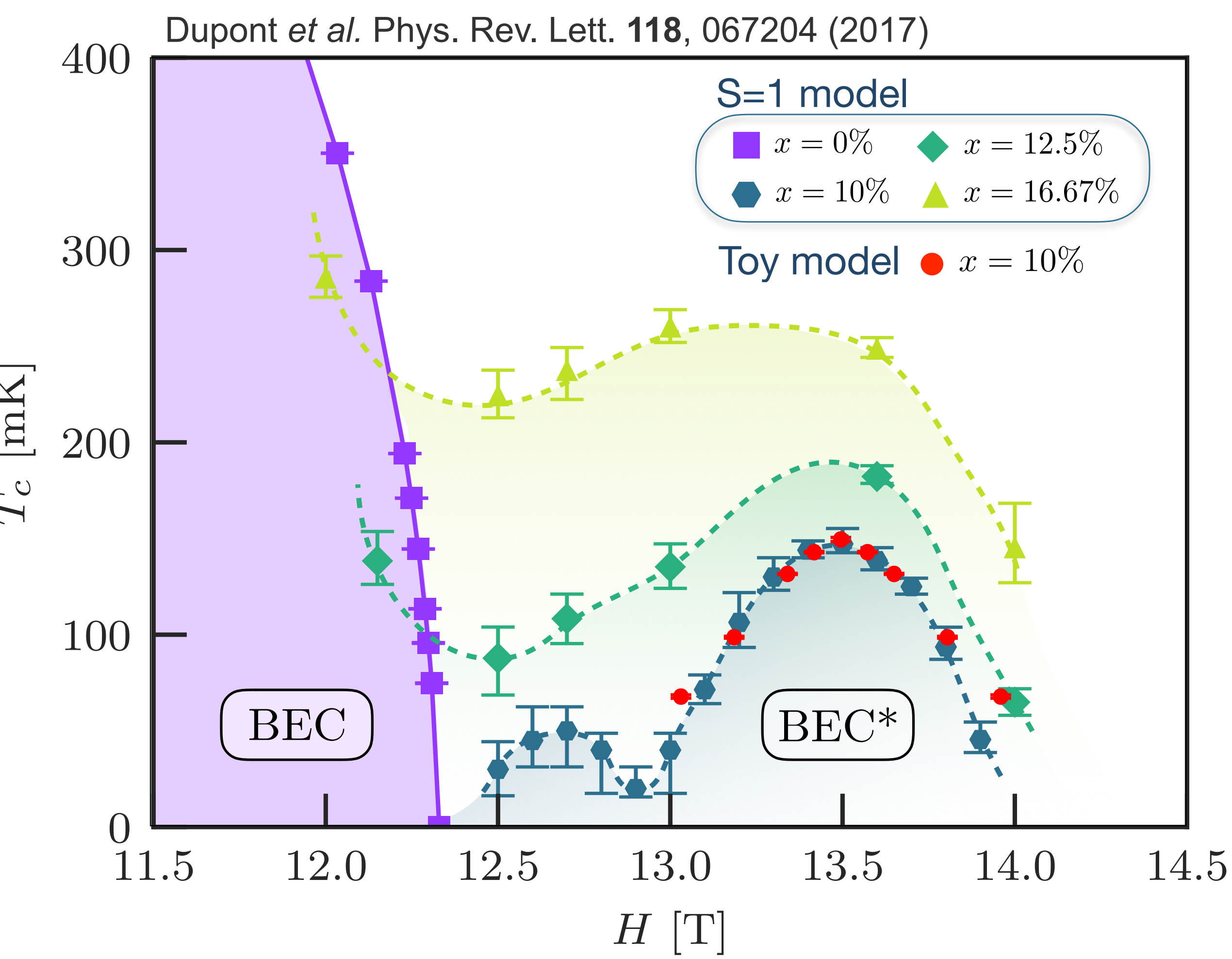
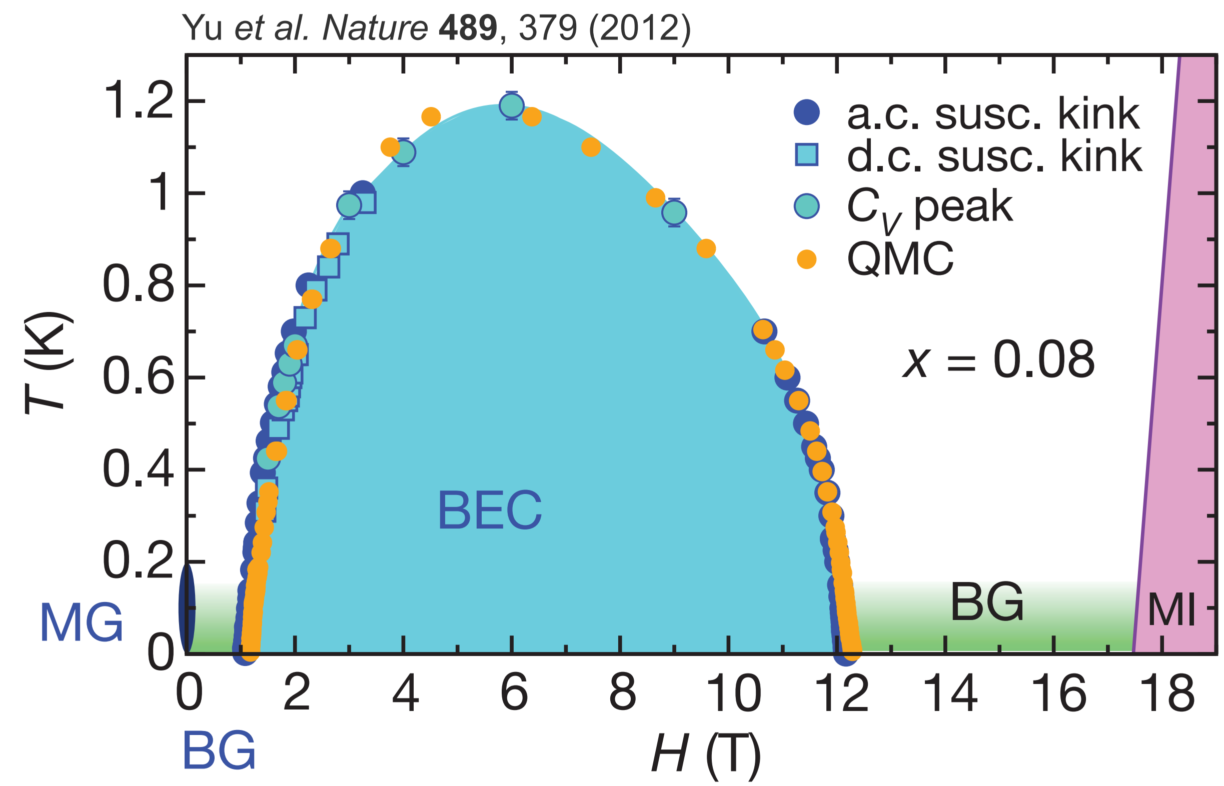
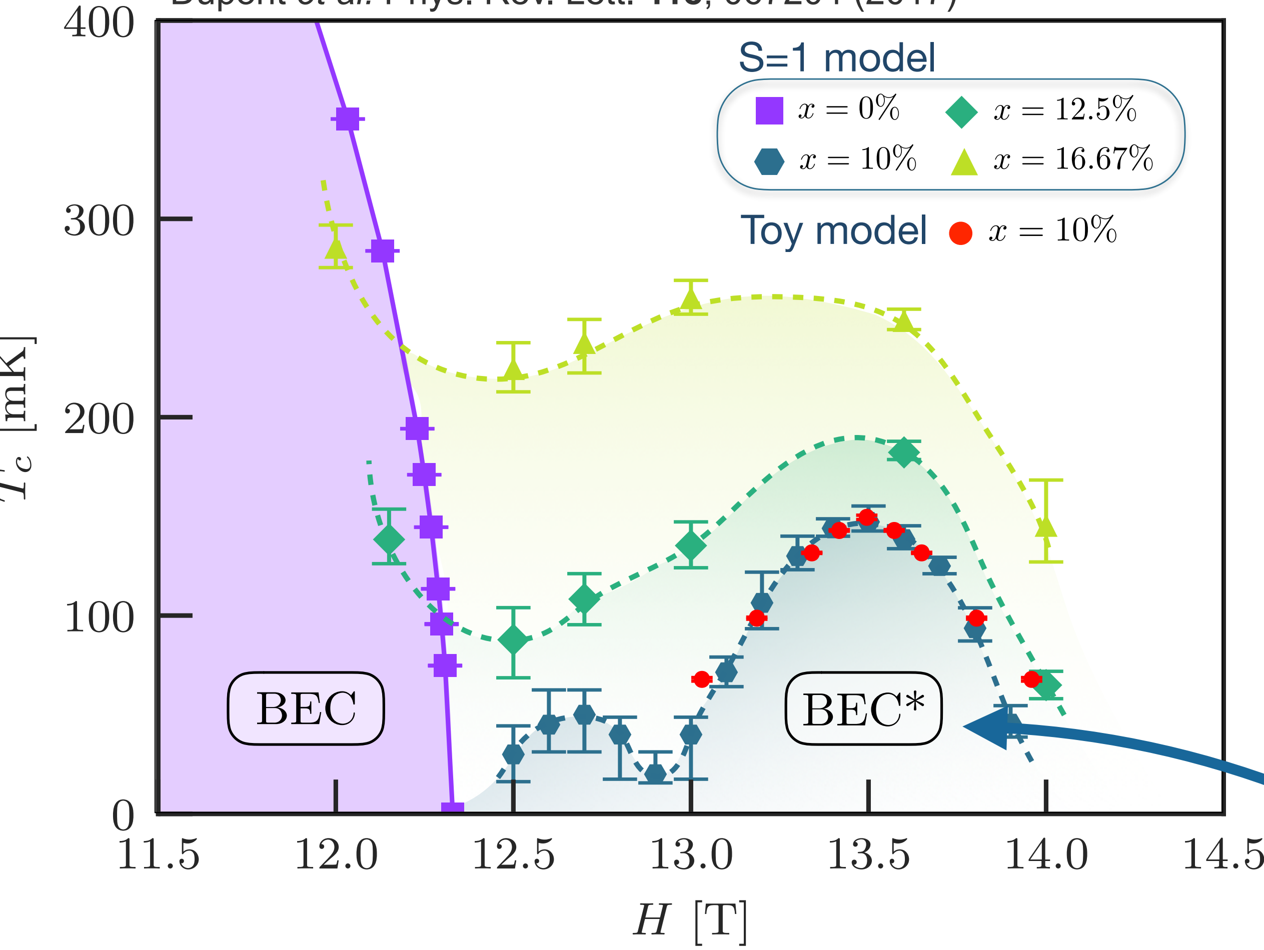


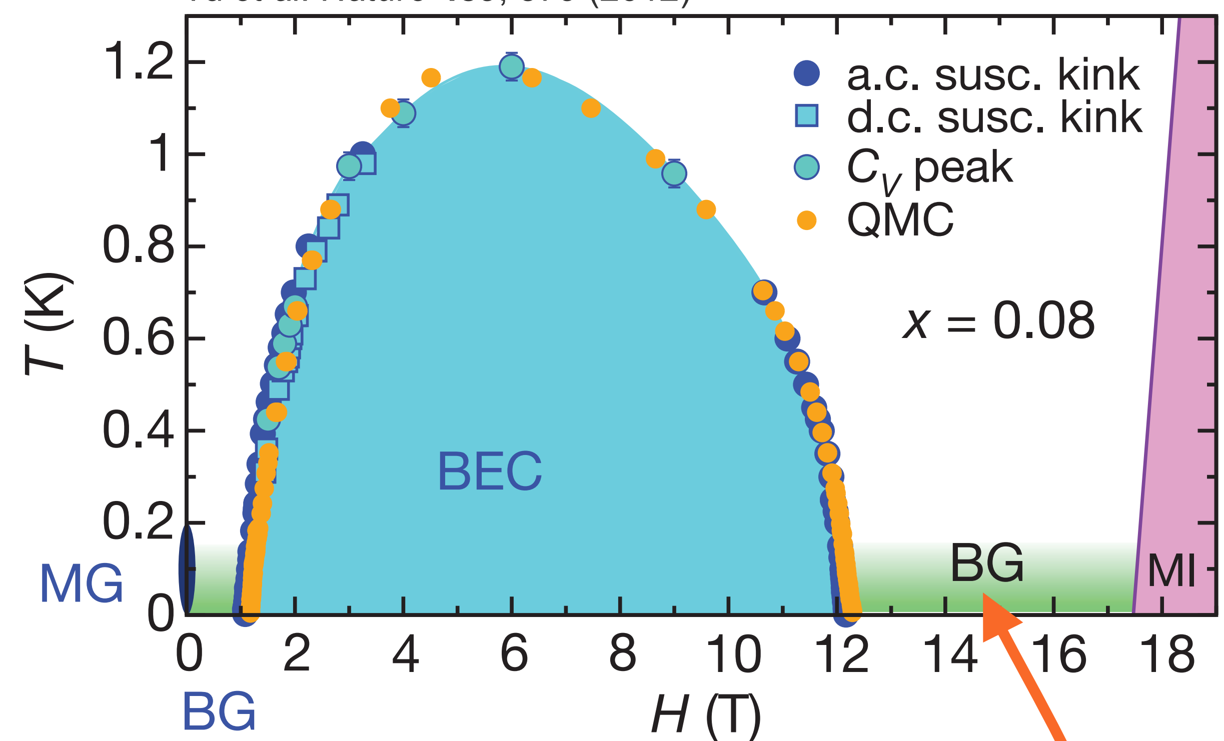


THEORY VS EXPERIMENTS

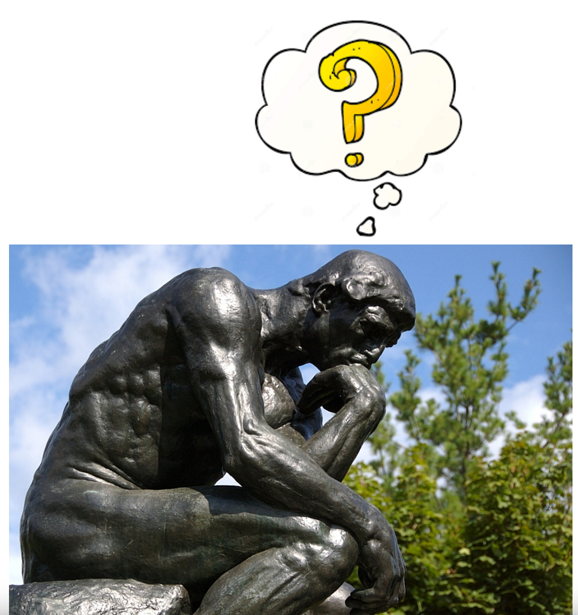
Dupont *et al.* Phys. Rev. Lett. **118**, 067204 (2017)



Yu *et al.* *Nature* **489**, 379 (2012)



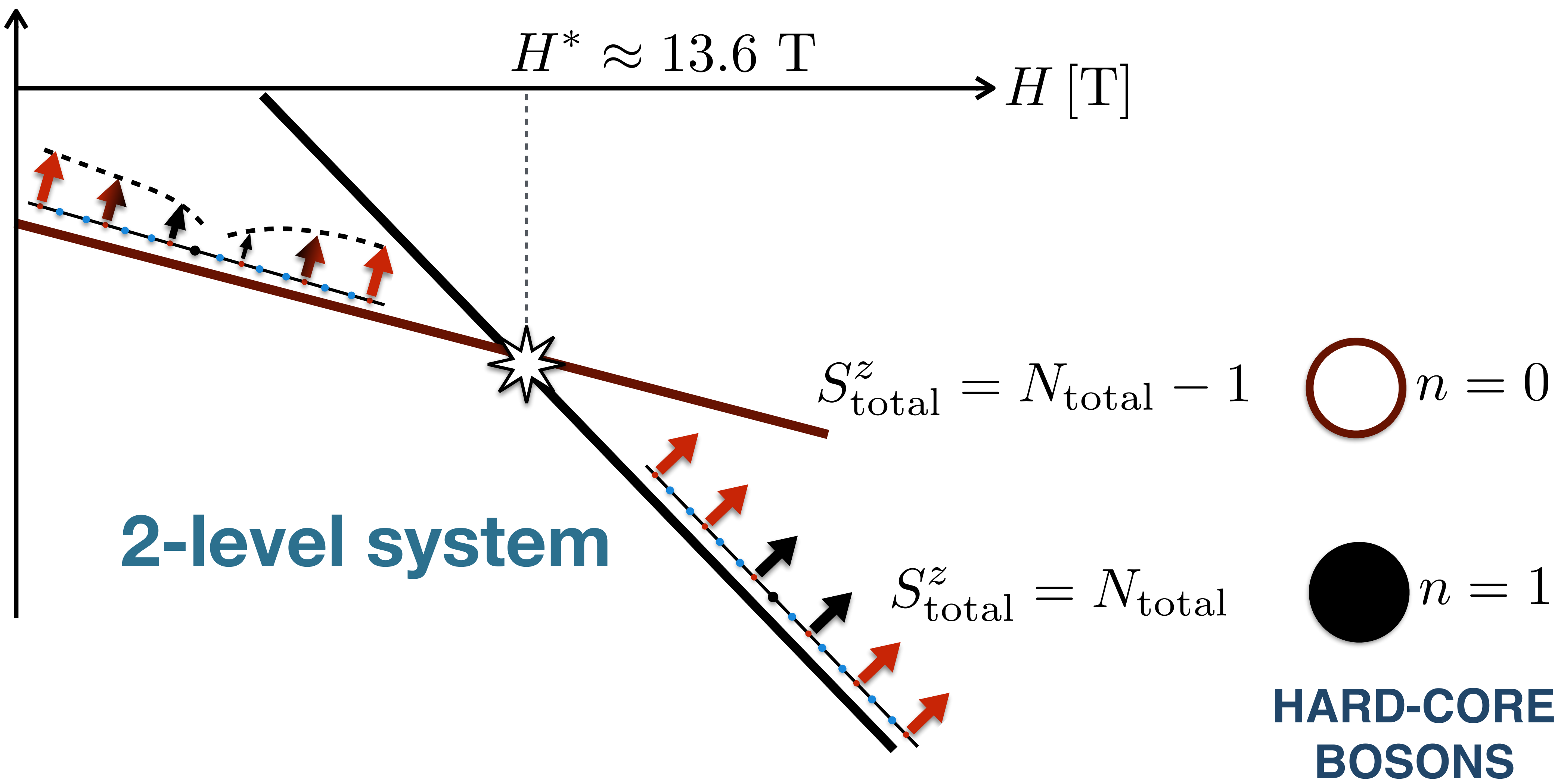
1) What is this new ordered phase ?



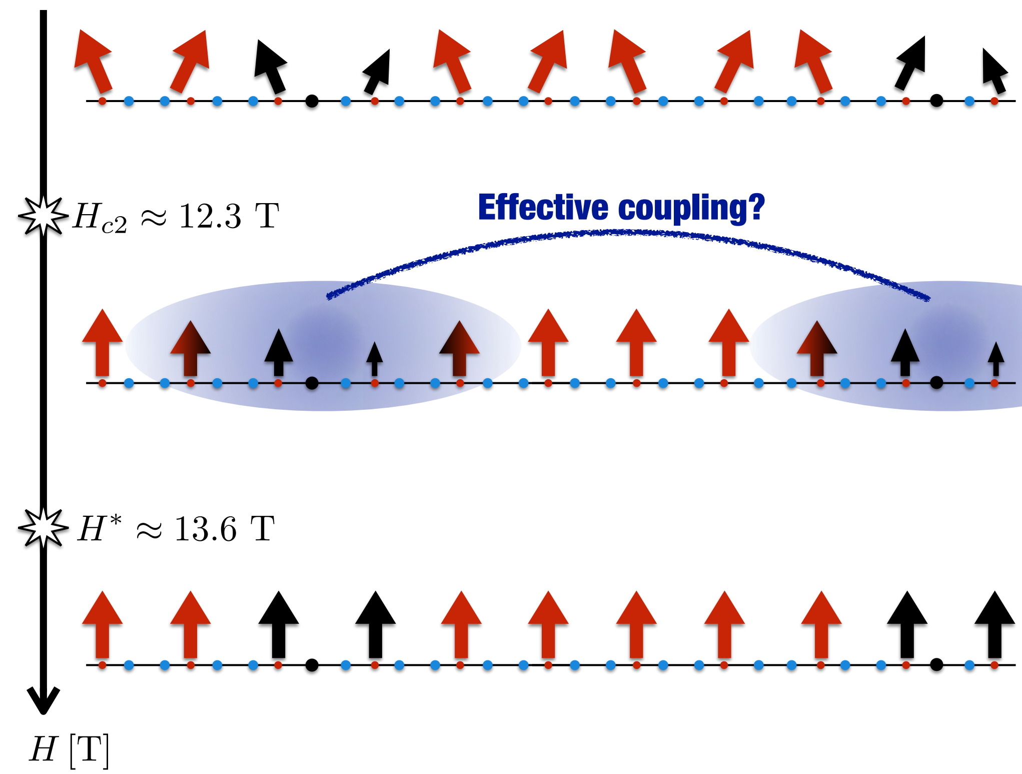
2) Where did the Bose glass slip away ??

3) Is BEC* real (experimentally) ???

LOCALIZED IMPURITY STATES BEHAVE LIKE HARD-CORE BOSONS



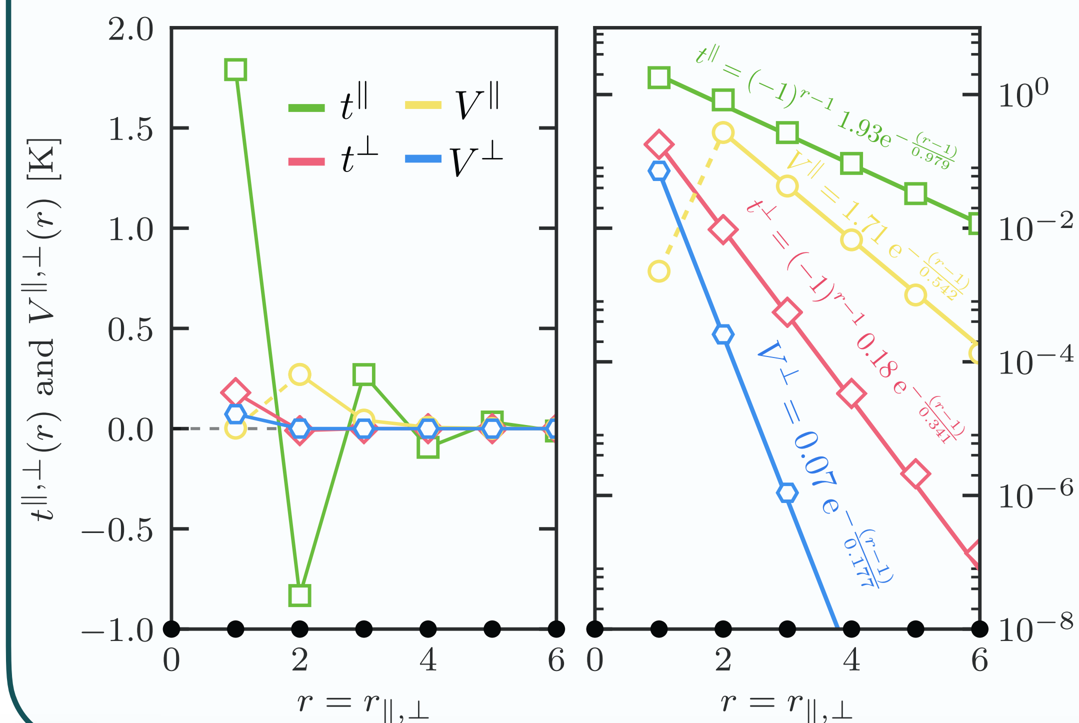
BEYOND SINGLE IMPURITY: HARD CORE BOSONS INTERACT!



BEYOND SINGLE IMPURITY: HARD CORE BOSONS INTERACT!

- Exact diagonalization with 2 impurities

Effective interactions

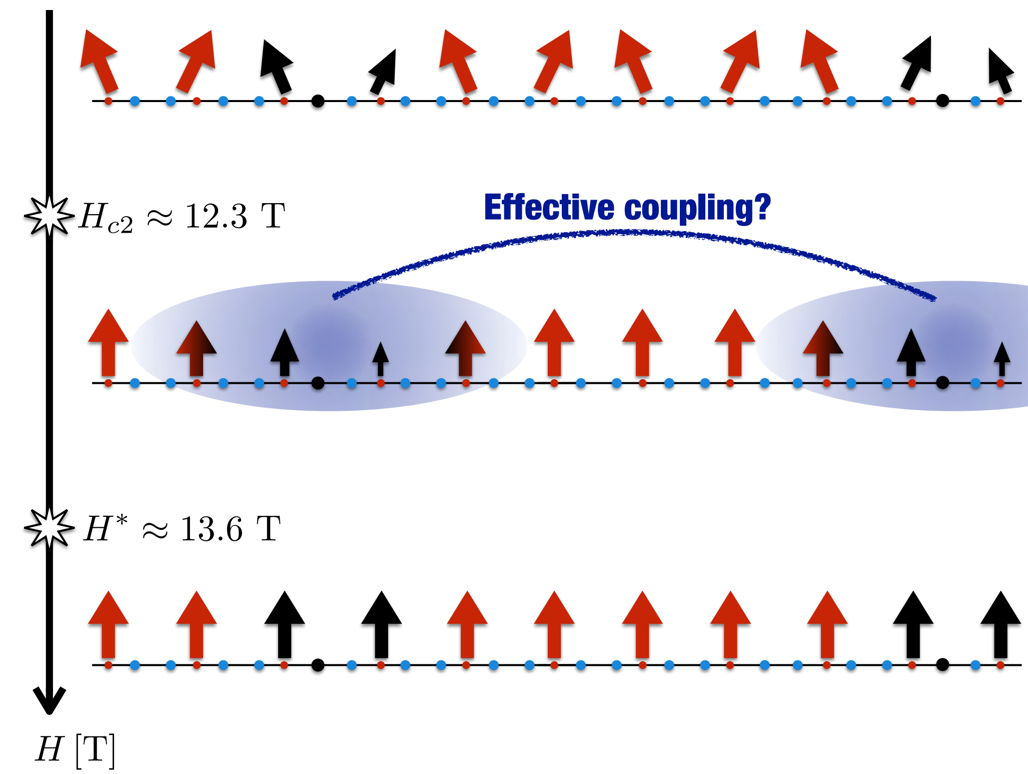


➡ Dominated by hoppings

➡ Non-frustrated

➡ Exponentially suppressed

$$|t_{ij}| \sim \exp(-r_{ij}/\xi)$$



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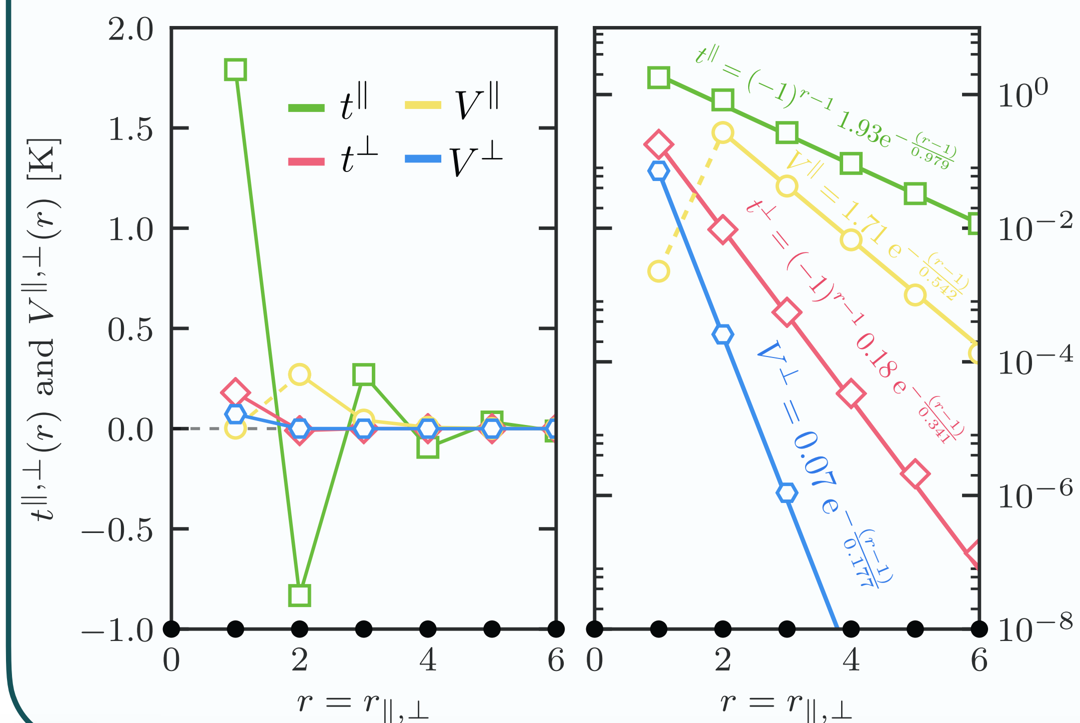

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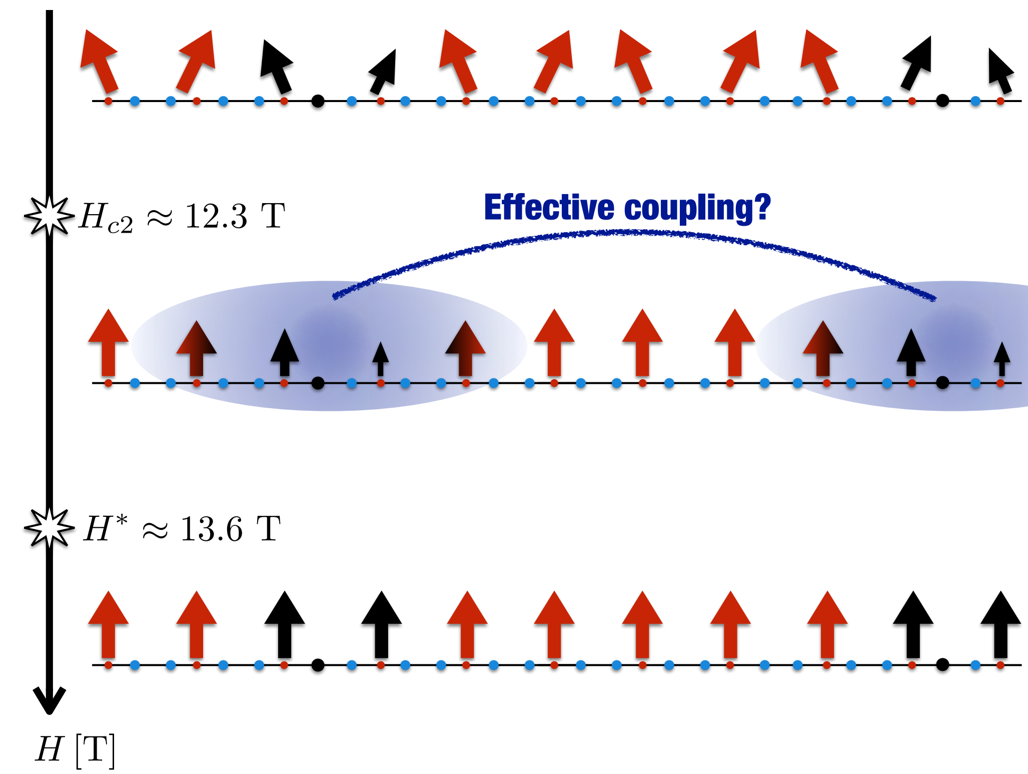

- Many impurities: Effective (hard-core) bosonic model for the impurity states

$$\mathcal{H}_{\text{eff}} = -\sum_{i,j} t_{ij} \left(b_i^\dagger b_j + \text{h.c.} \right) - (H - H^*) \sum_i n_i$$

<u>Random hoppings</u>
due to random positions
of impurities

<u>chemical potential</u>
controls the density
of hard-core bosons

These objects may acquire a 3D coherence in the vicinity of half-filling for H~H* => The BEC* phase!

IS THE BEC* PHASE OBSERVABLE….?

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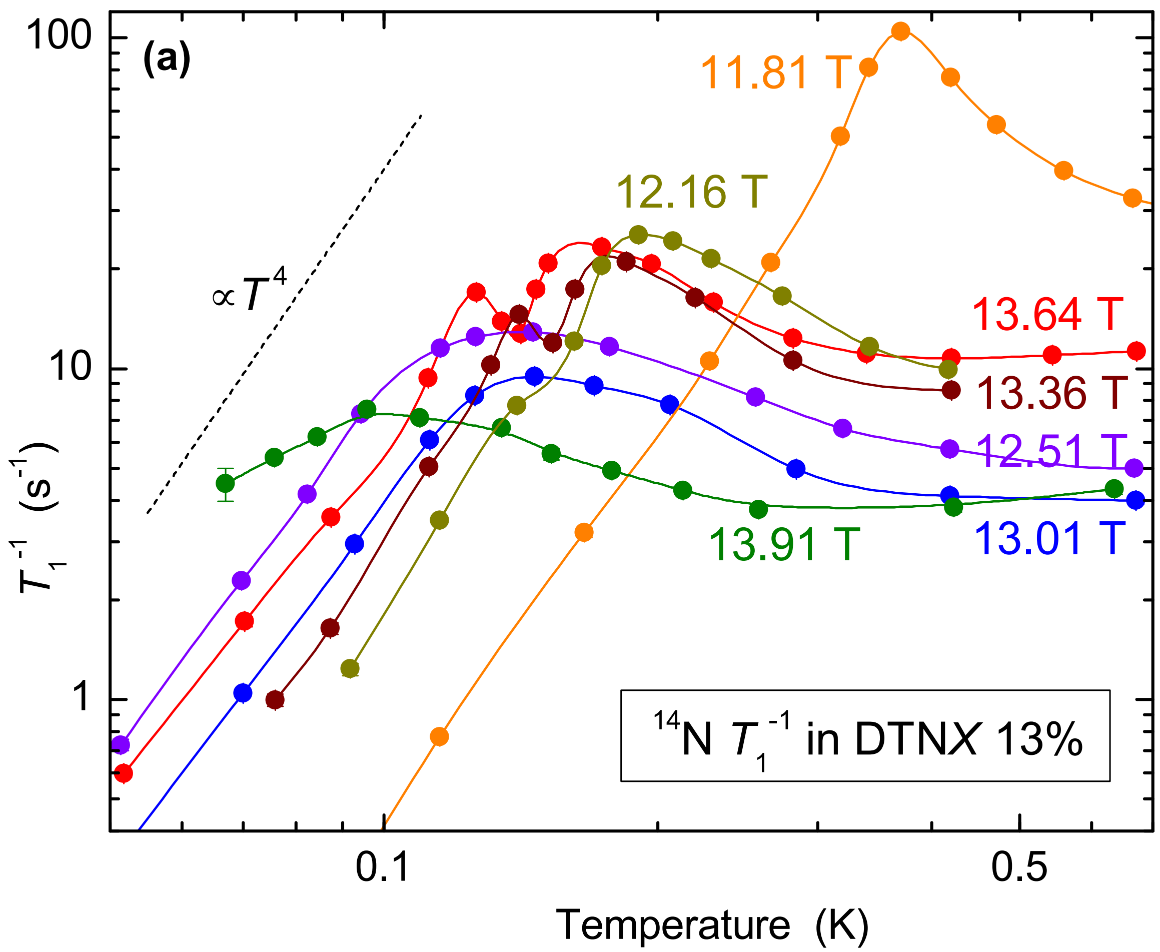
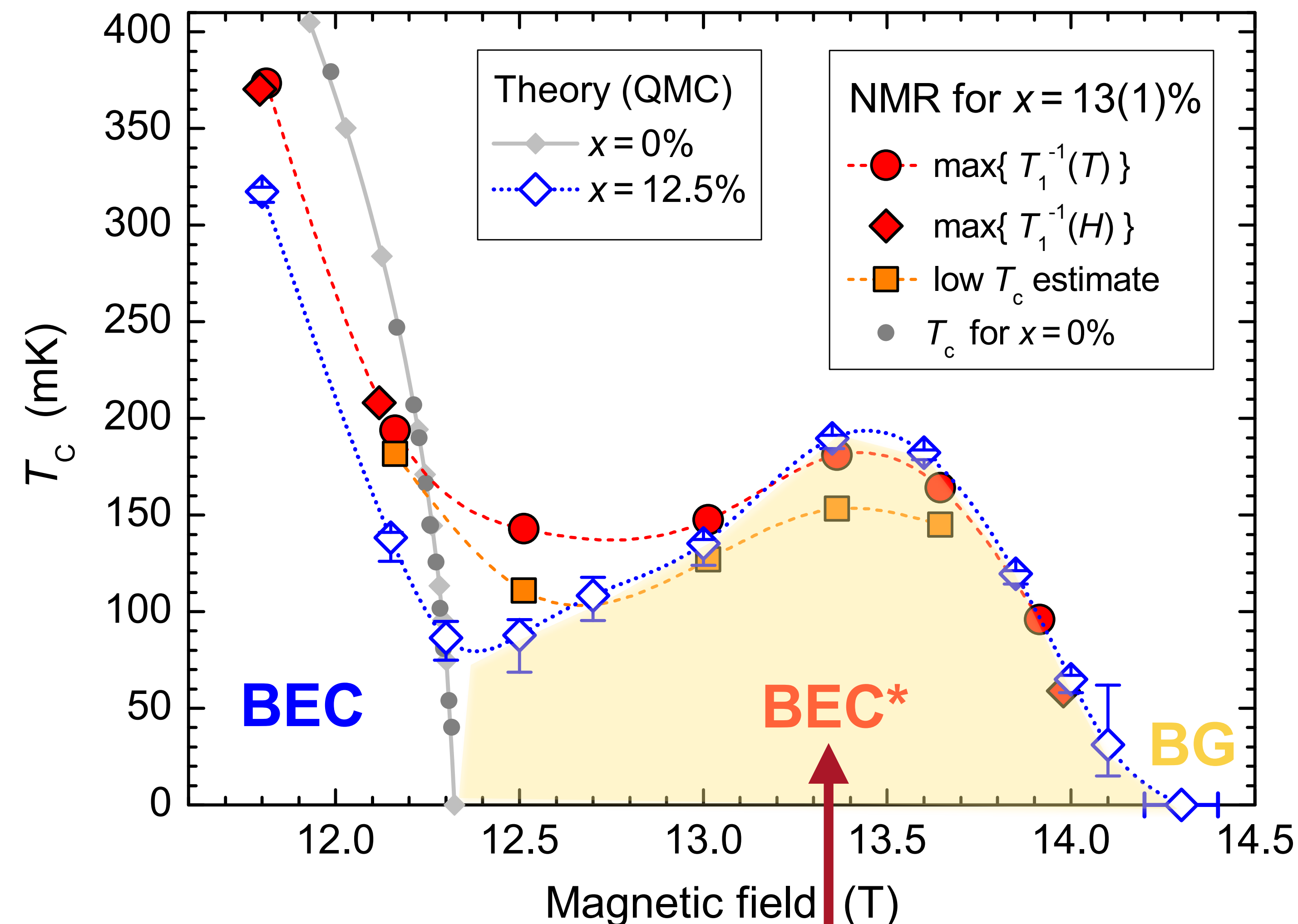
👍 A. Orlova *et al.*
PRL 2018

IS THE BEC* PHASE OBSERVABLE…?

A. Orlova *et al.*
PRL 2018

- NMR relaxation



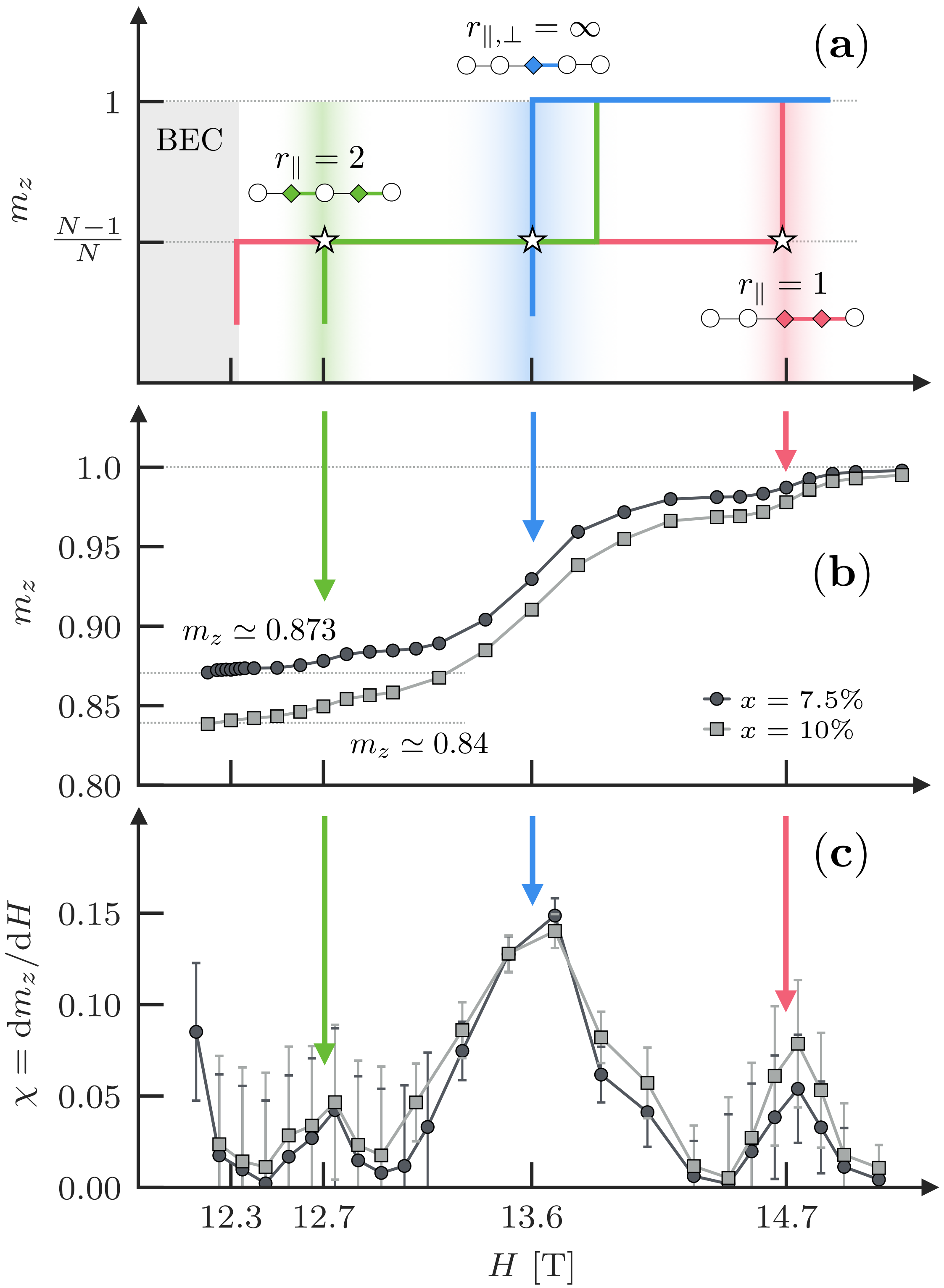


Field- <u>and</u> Disorder-induced Ordered (BEC*) phase

MORE ?

M. Dupont *et al.* PRB 2017

◉ Multi-impurity effects



MORE ?

M. Dupont *et al.* PRB 2017

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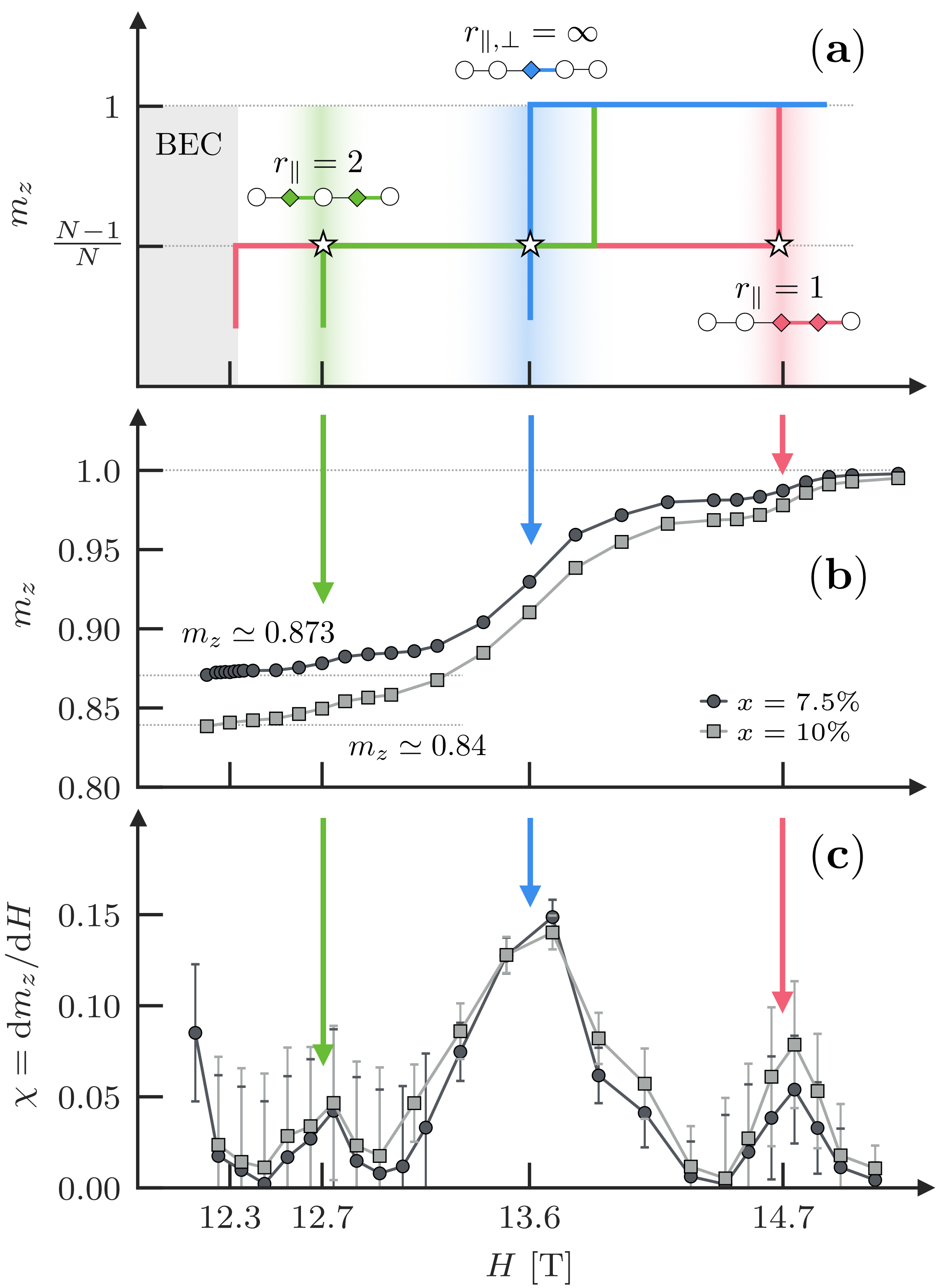

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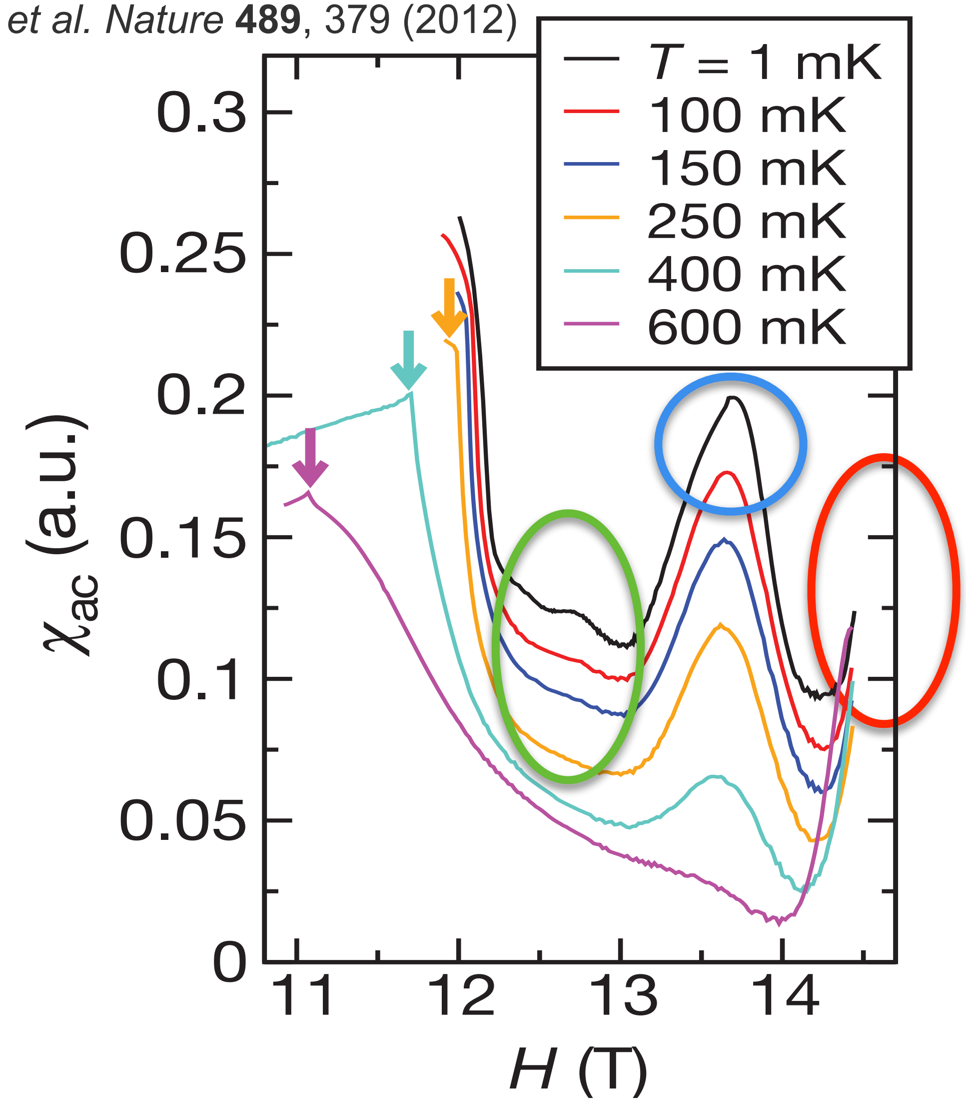

MORE ?

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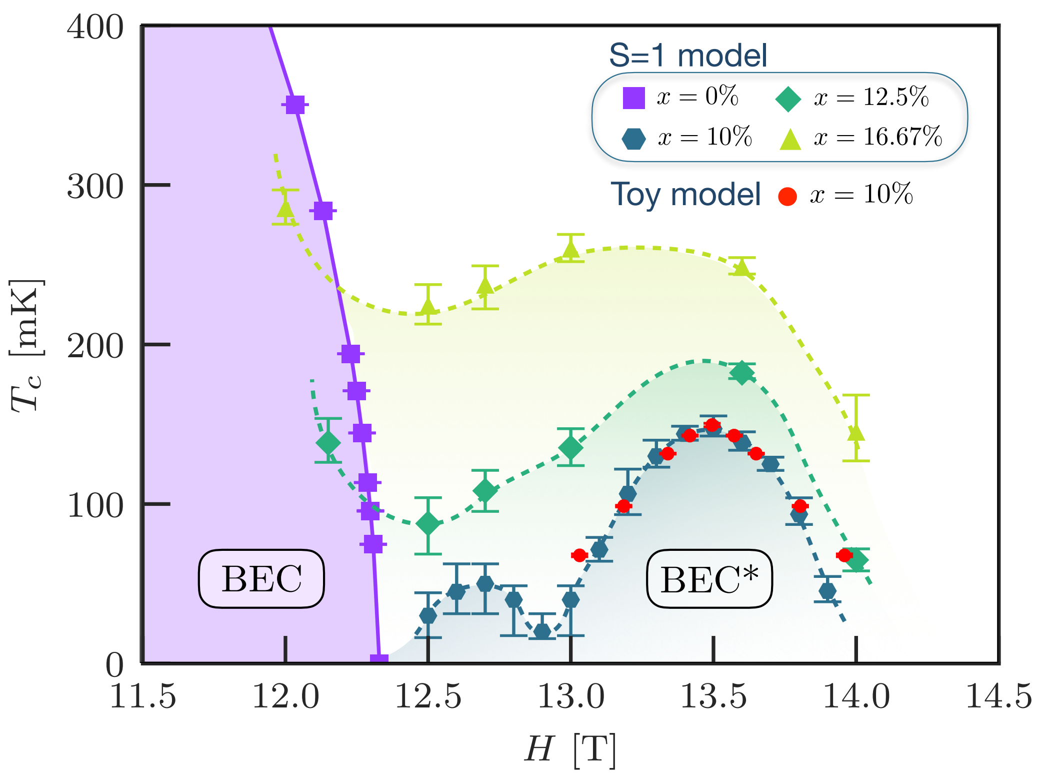

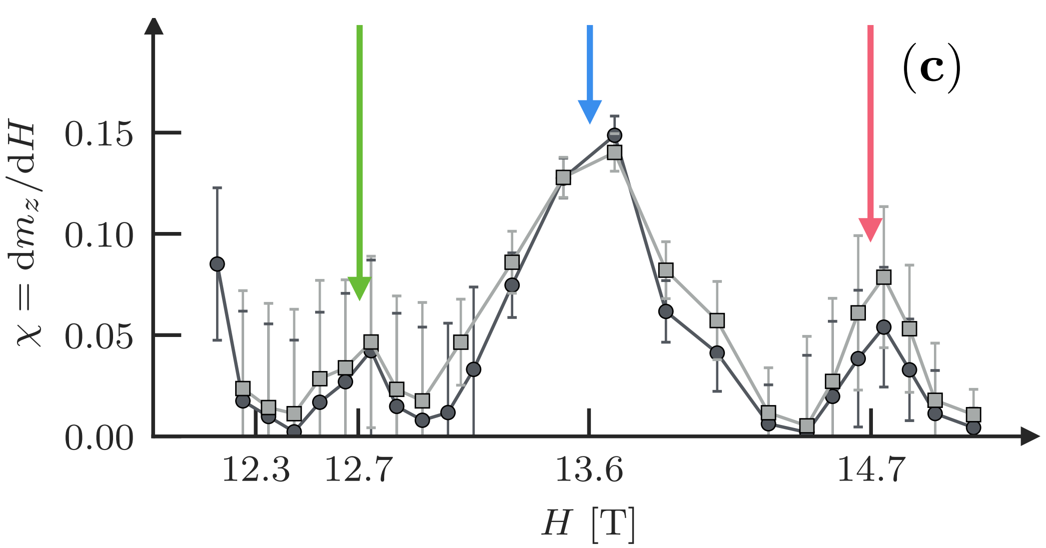

MORE ?

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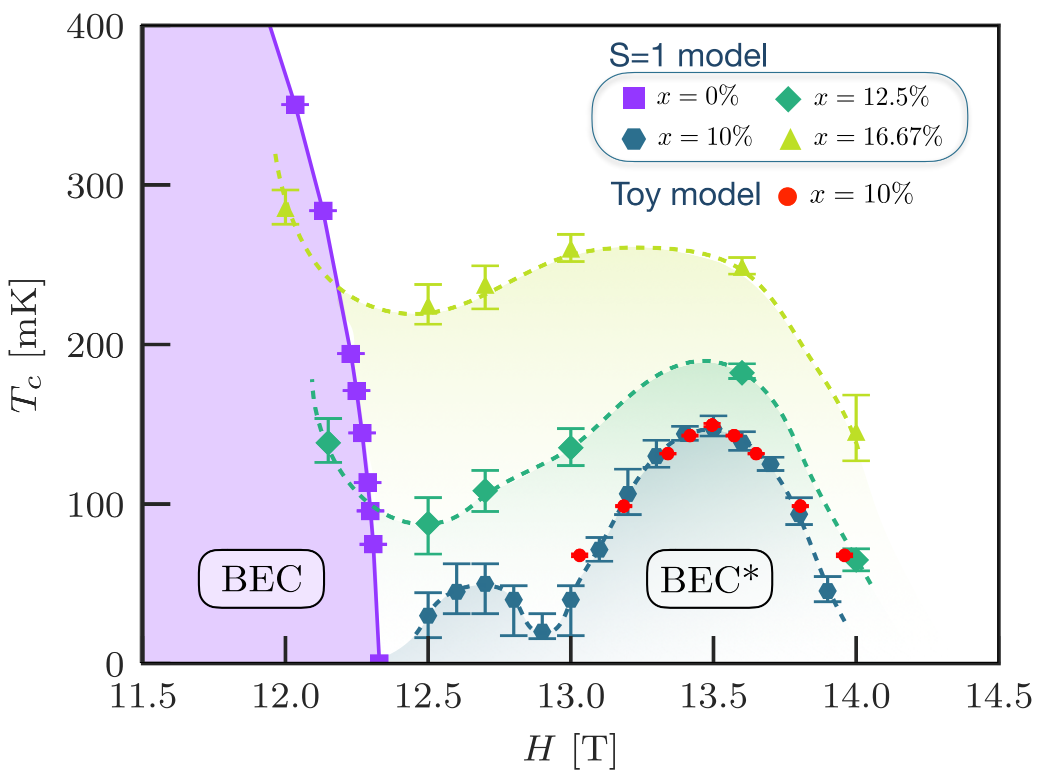
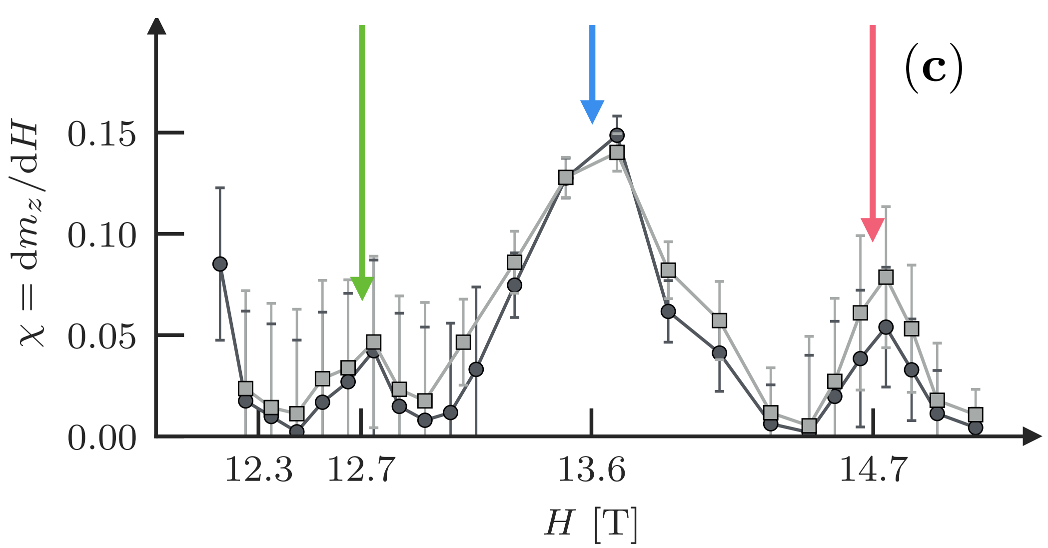
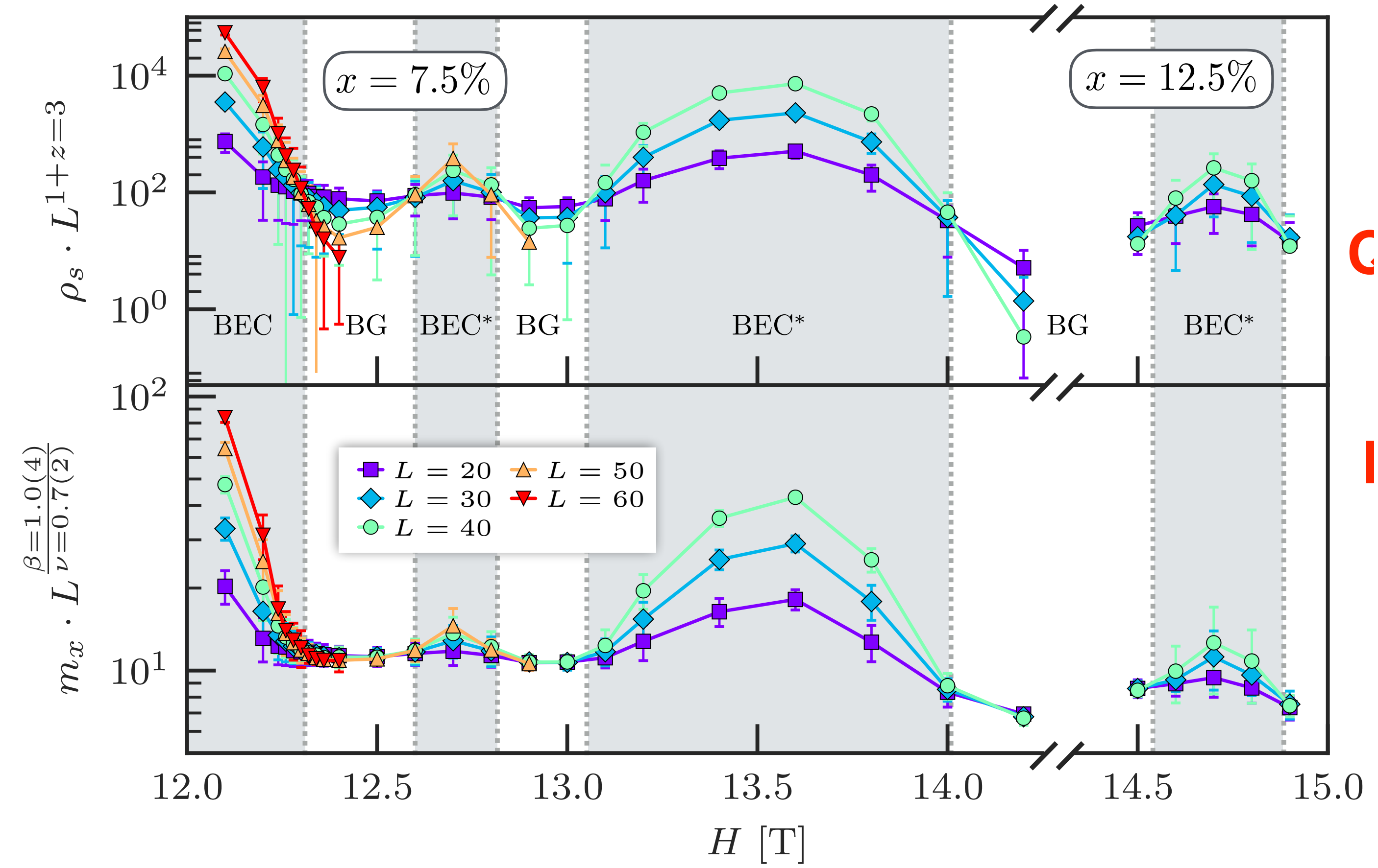




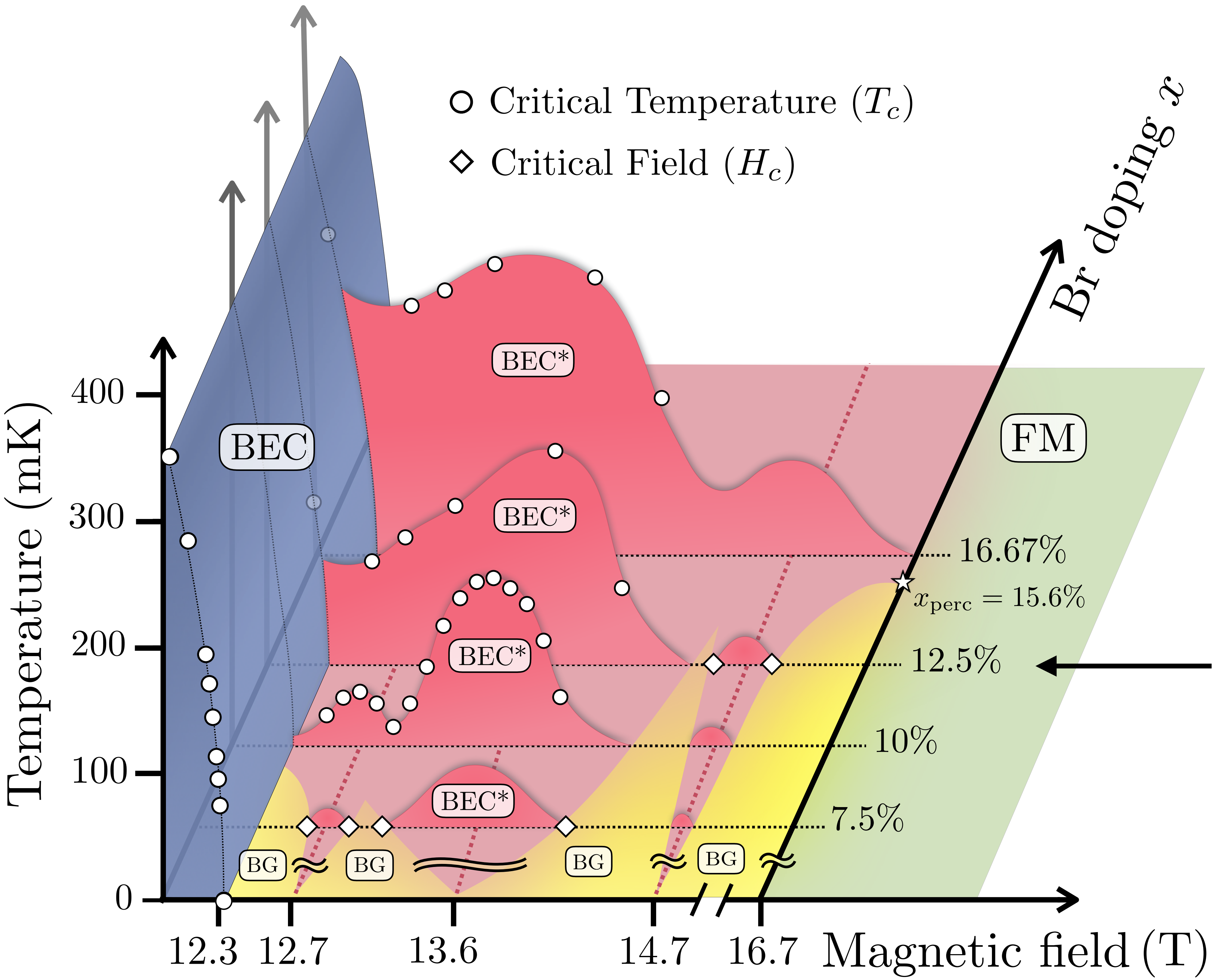
◉ $T = 0$ Bose glass?



We expect a succession of T=0 BEC - BG transitions

GLOBAL PHASE DIAGRAM

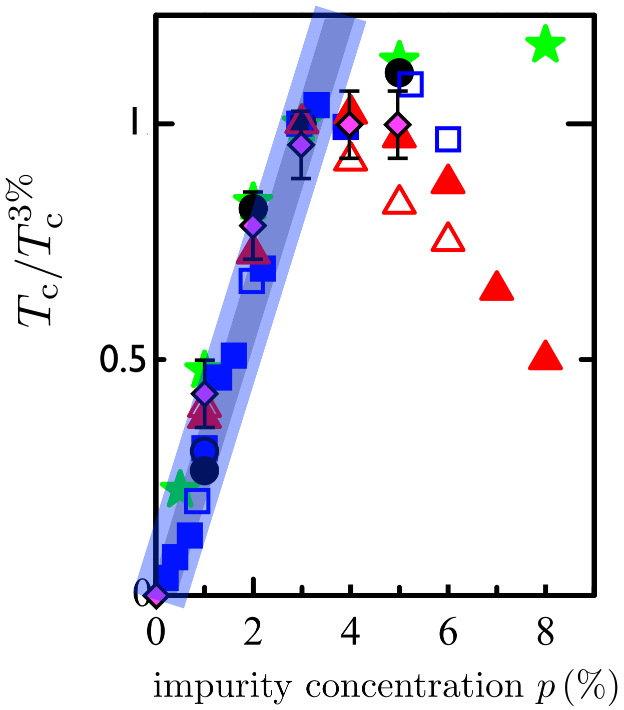
M. Dupont *et al.* PRB 2017



Our experiments

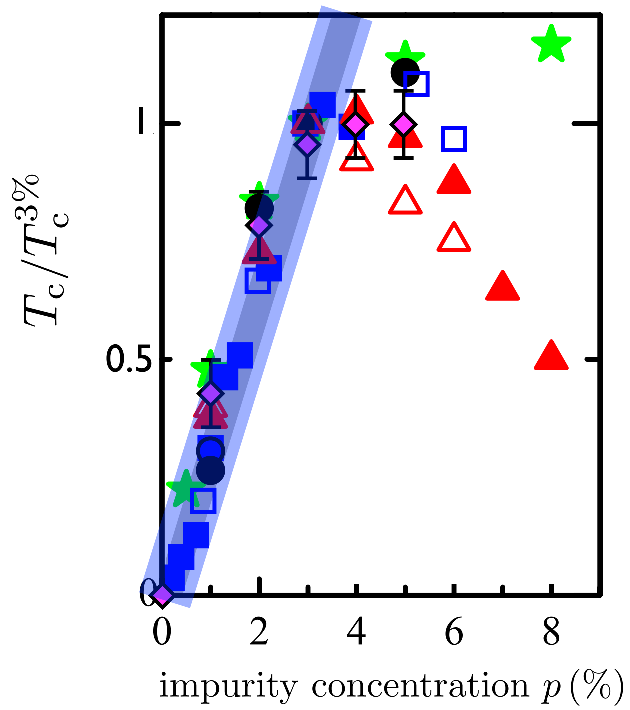
CONCLUSIONS

CONCLUSIONS



– **Zero-field spin gapped systems with impurities: generic impurity-induced "order-from-disorder" mechanism**

CONCLUSIONS



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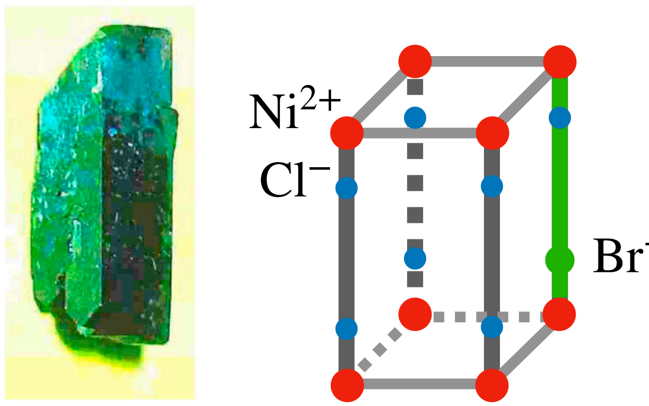
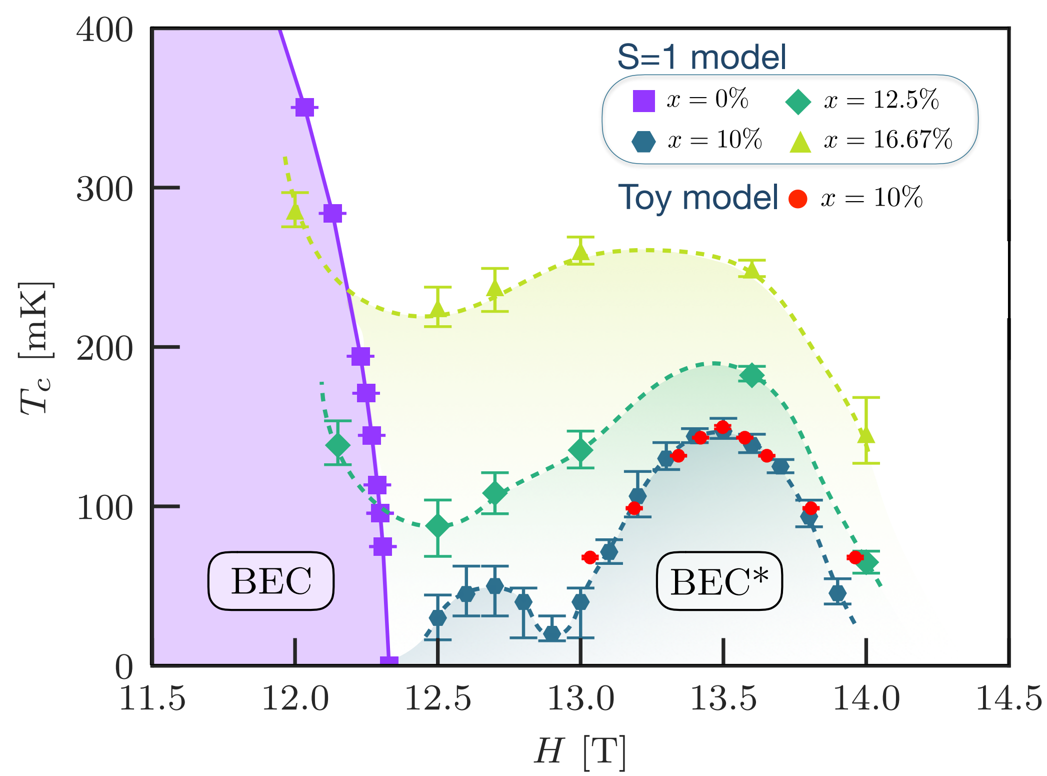
– DTNX material

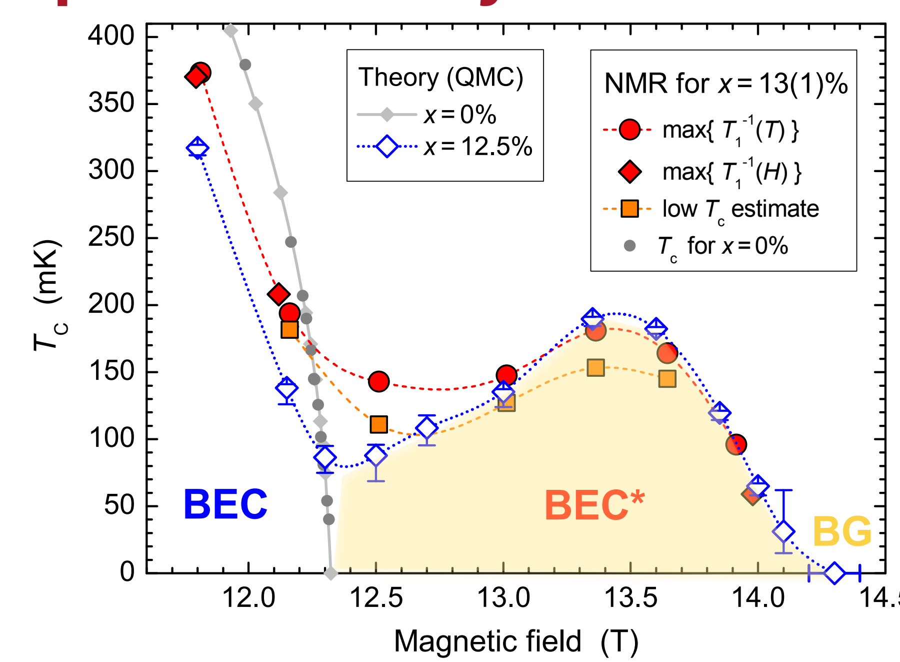
- microscopic modelling



- **Theoretical prediction of a new disorder and field-induced BEC* phase**

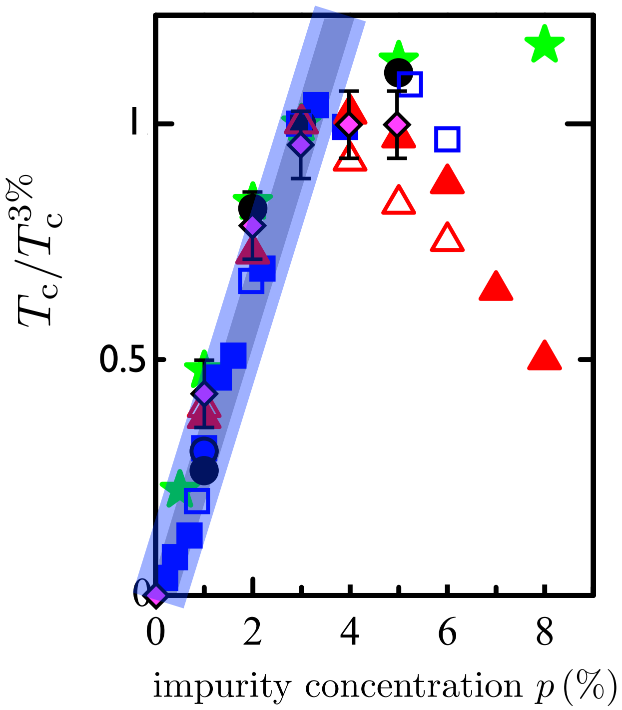


- **Experimentally confirmed**



Subtle cooperative manifestation of disorder AND interactions !

CONCLUSIONS



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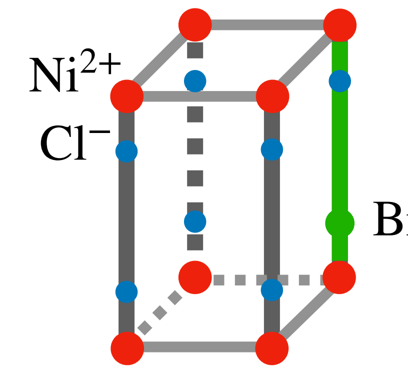

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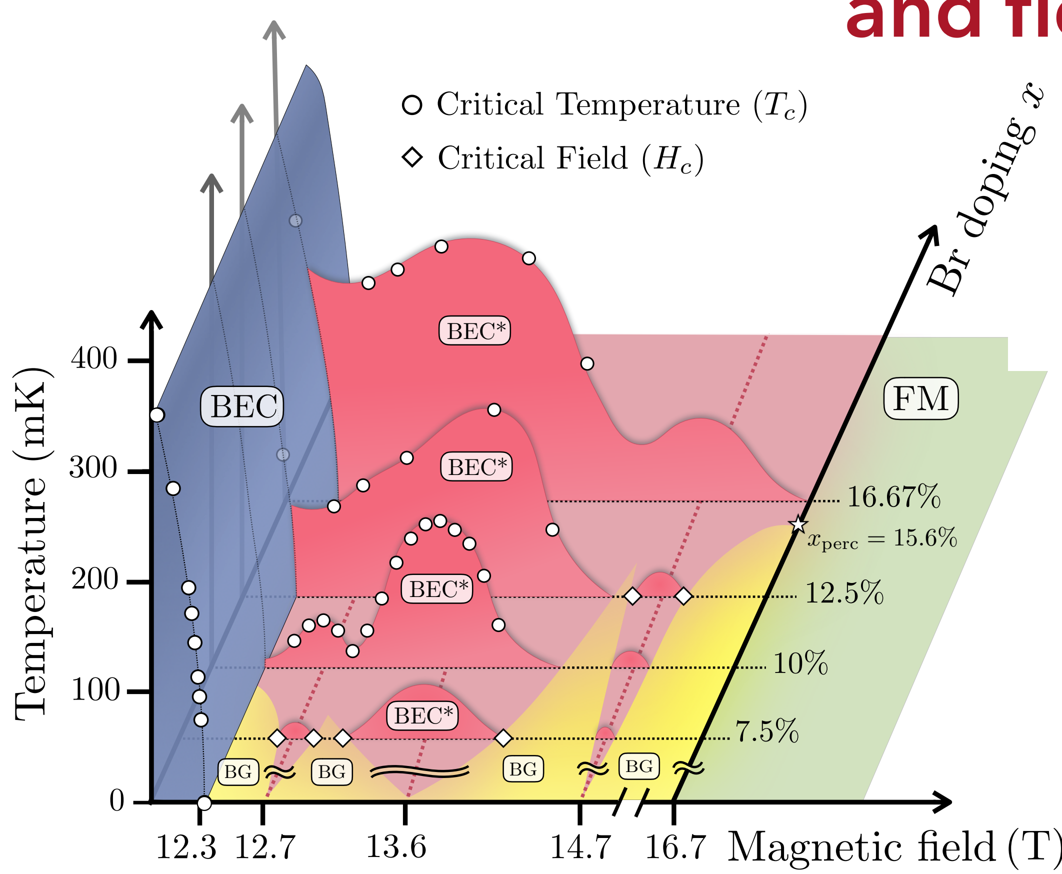

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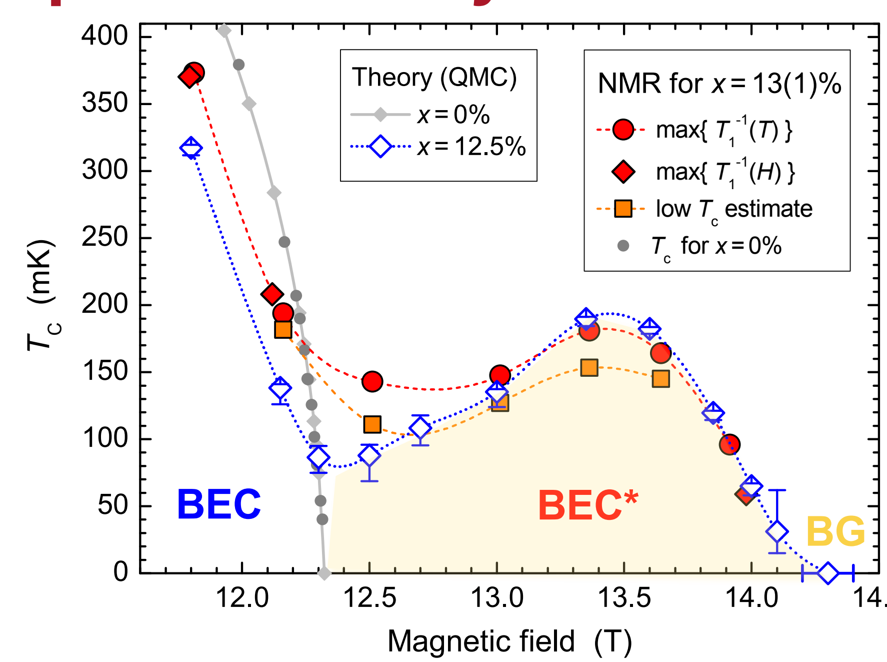

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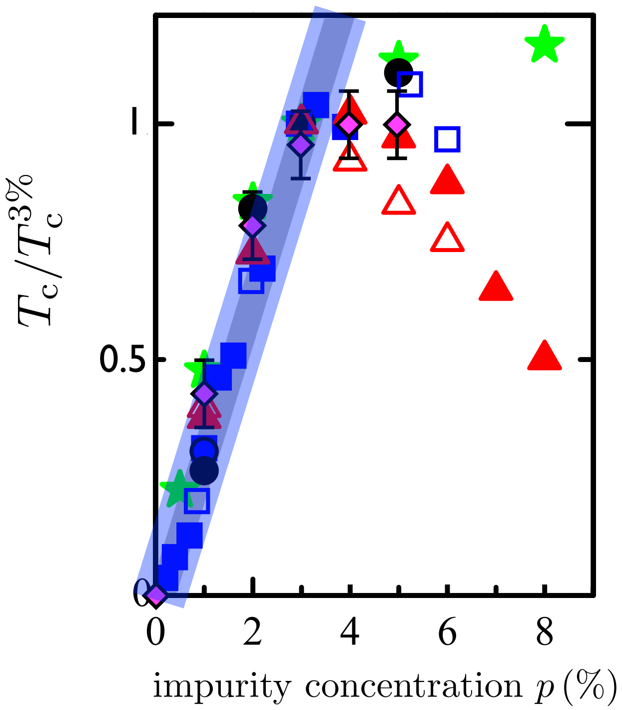
CONCLUSIONS



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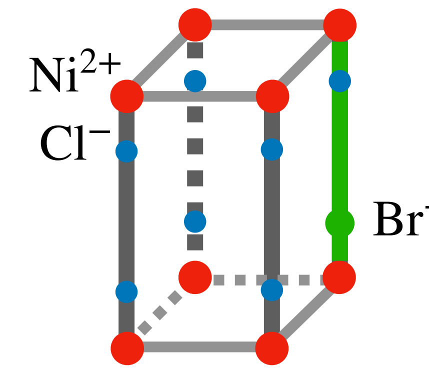

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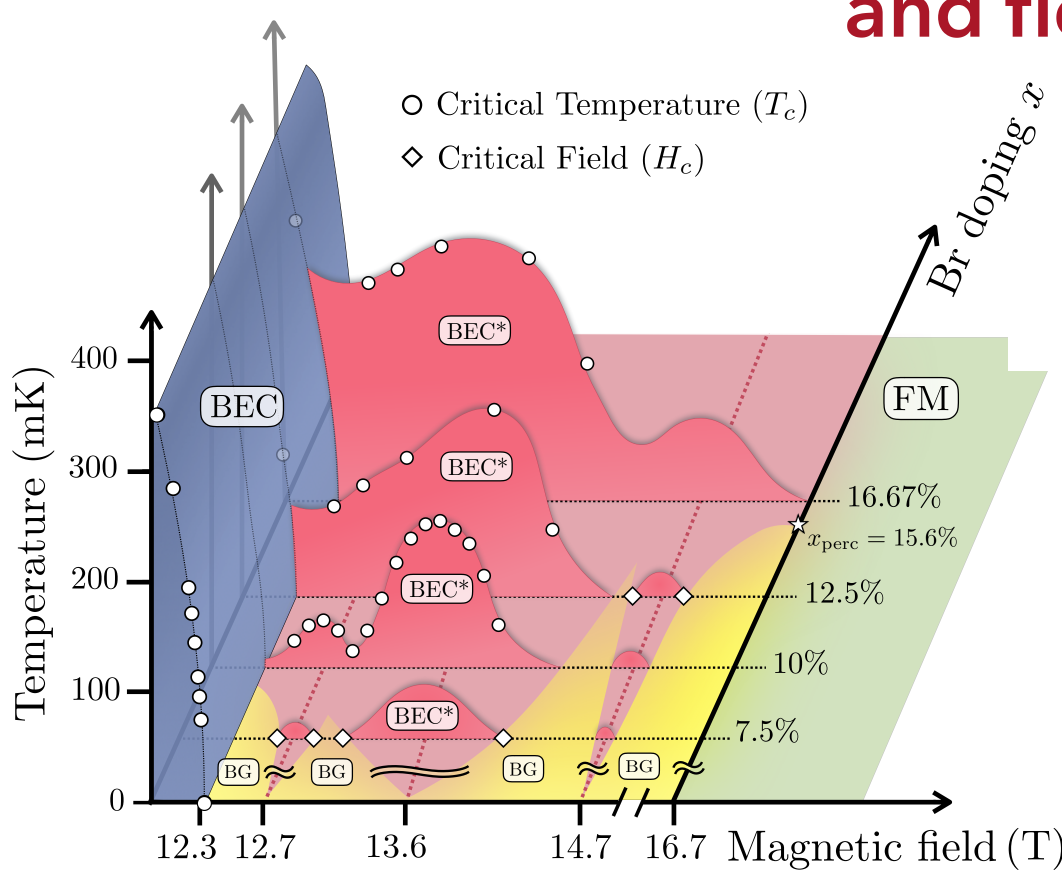

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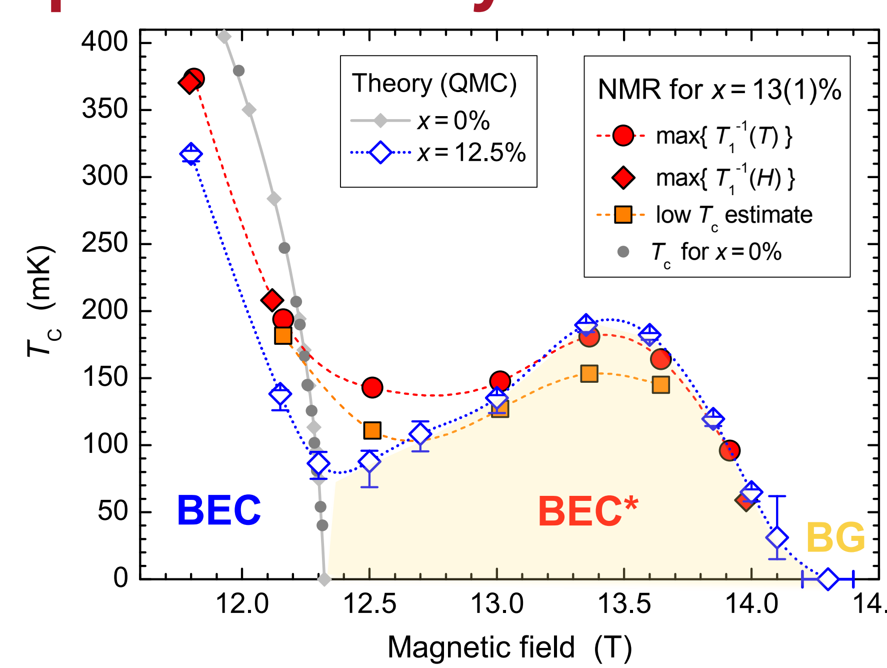

Subtle cooperative manifestation of disorder AND interactions !