# Small Angle Neutron Scattering Study of the nanometric phase separation in $Pr_{1-x}Ca_xMnO_3$

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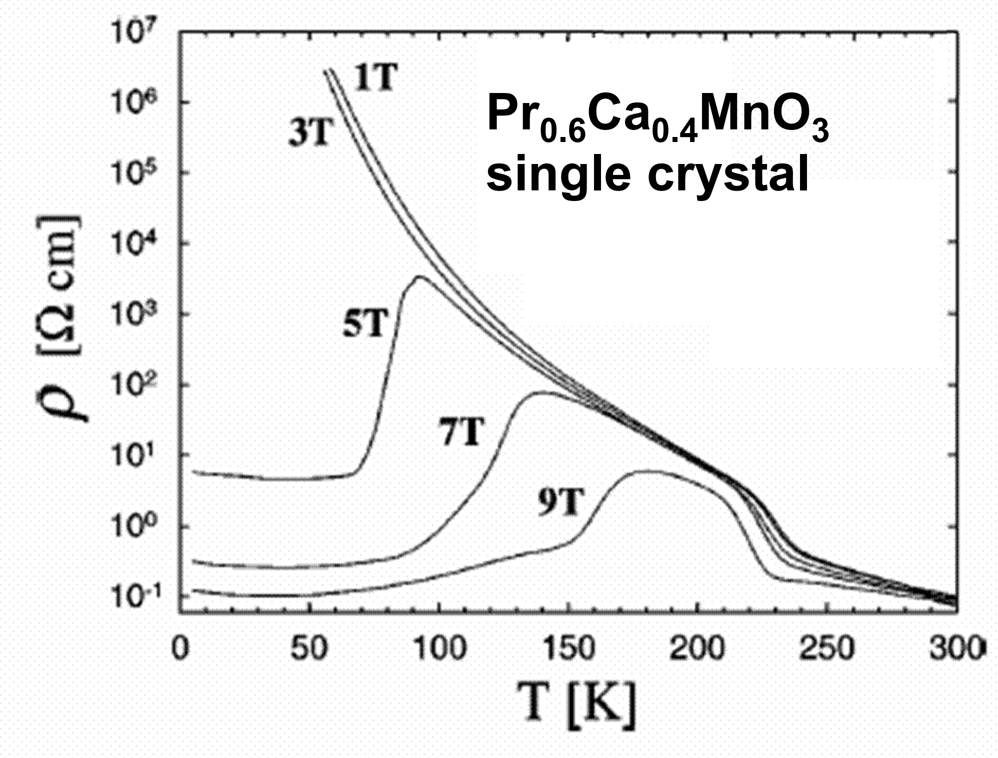
# WHY STUDYING THESE COMPOUNDS ?

## COLOSSAL MAGNETORESISTANCE (CMR) :



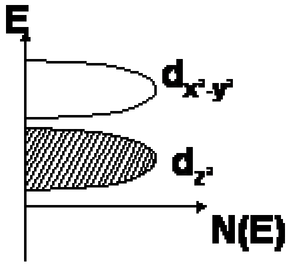
**$Pr_{0.6}Ca_{0.4}MnO_3$ single crystal**

*V. Hardy et al. , Phys. Rev. B 64, 64402 (2001)*

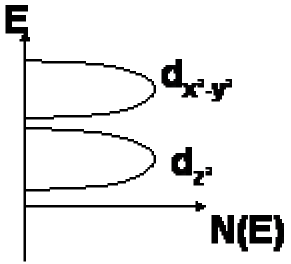
# A COMPETITION BETWEEN TWO EXCHANGE MECHANISMS :

$Pr^{(3+)}_{(1-x)}\ Ca^{(2+)}_{(x)}Mn^{(3+)}_{(1-x)}\ Mn^{(4+)}_{x}\ O^{(2-)}_{3}$

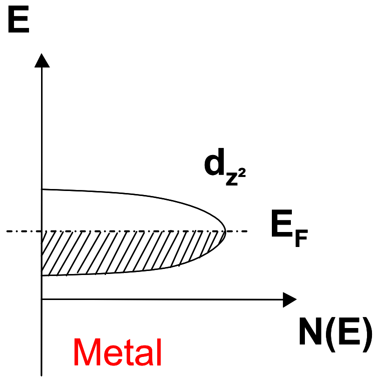
**$Mn^{3+}$**



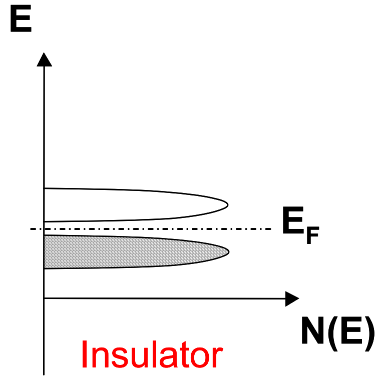
**$Mn^{4+}$**



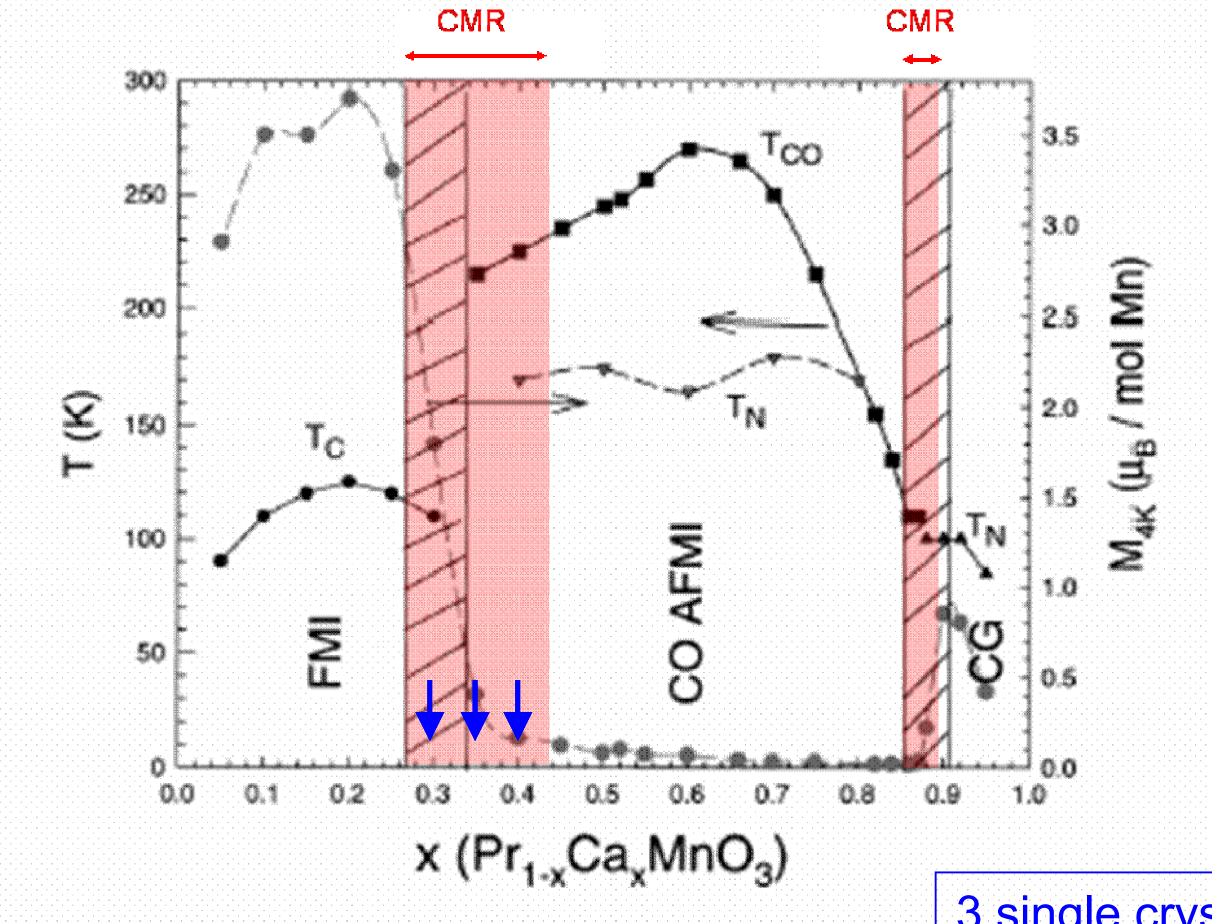
Delocalized charges
Ferromagnetism



Localized charges
Antiferromagnetism



# PHASE DIAGRAM OF THE $Pr_{1-x}Ca_xMnO_3$ SERIES :

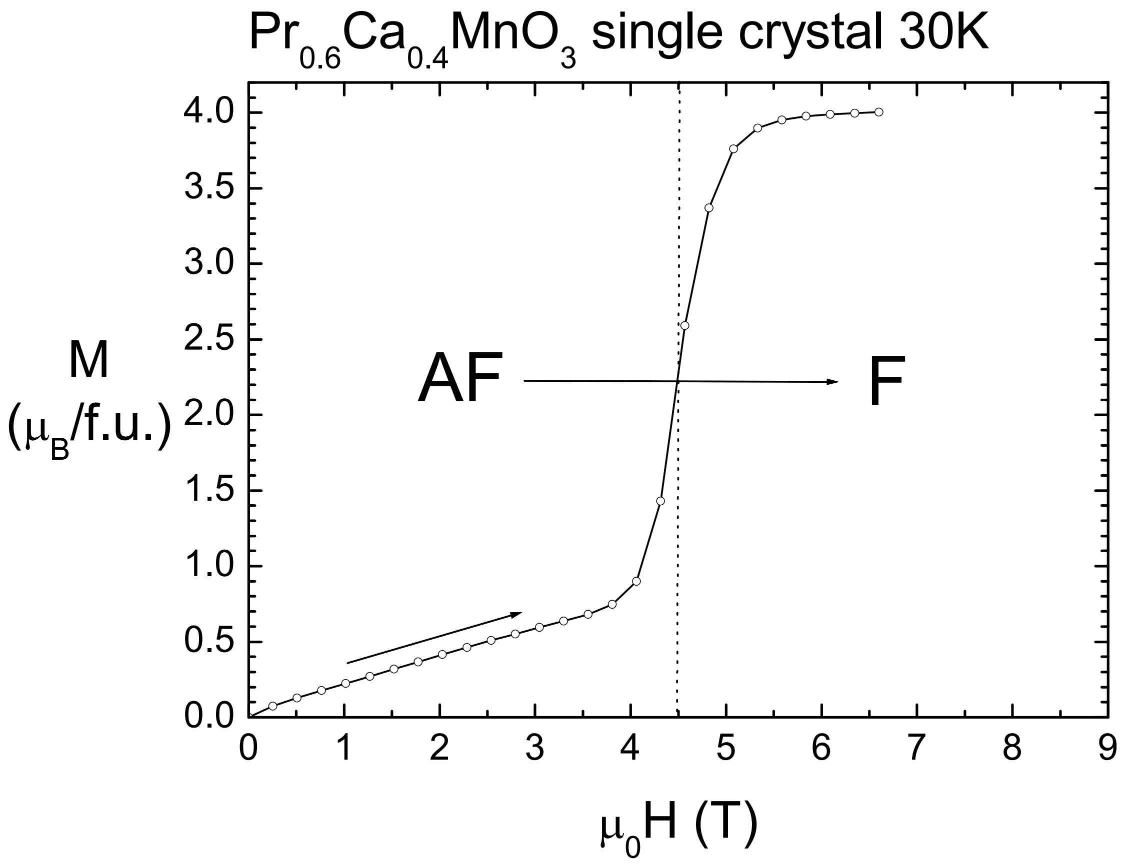


*C. Martin et al., Phys. Rev. B 60, 12191 (1999)*

3 single crystals:

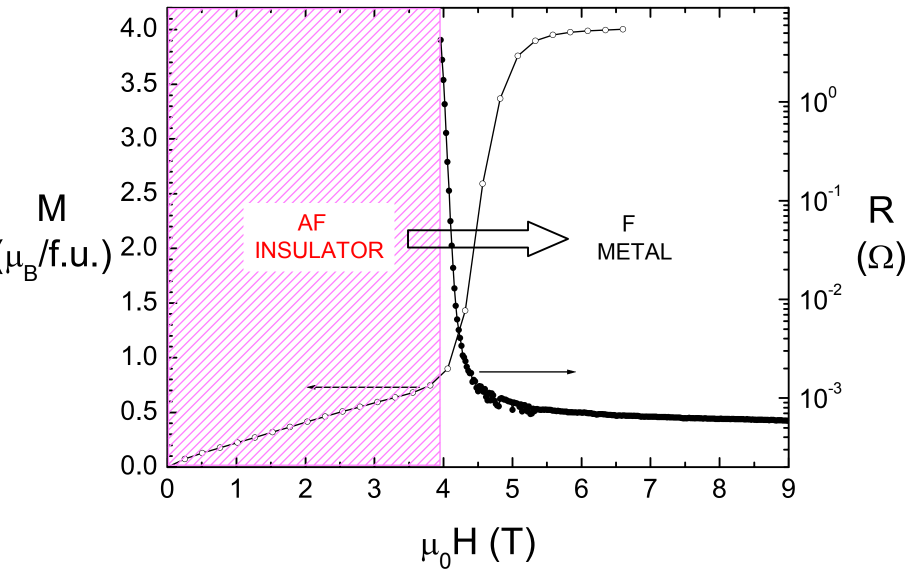
x=0.3, 0.35 and 0.4

# THE METAMAGNETIC TRANSITION :

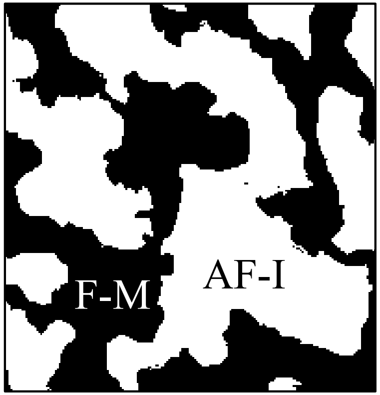


# THE INSULATOR-to-METAL TRANSITION AND THE PHASE SEPARATION :

$Pr_{0.6}Ca_{0.4}MnO_3$ single crystal 30K



PERCOLATION IN AN ELECTRONIC PHASE SEPARATED SYSTEM :



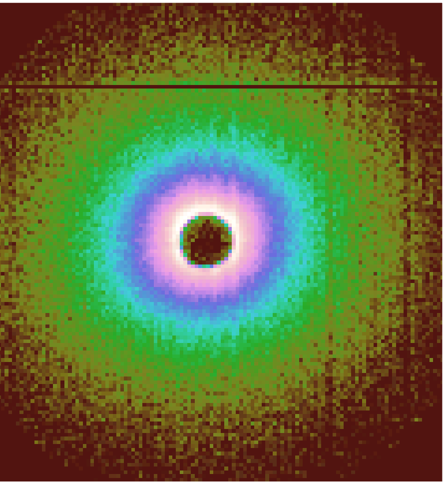
$$R \propto \left(\phi - \phi_T\right)^{-S}$$

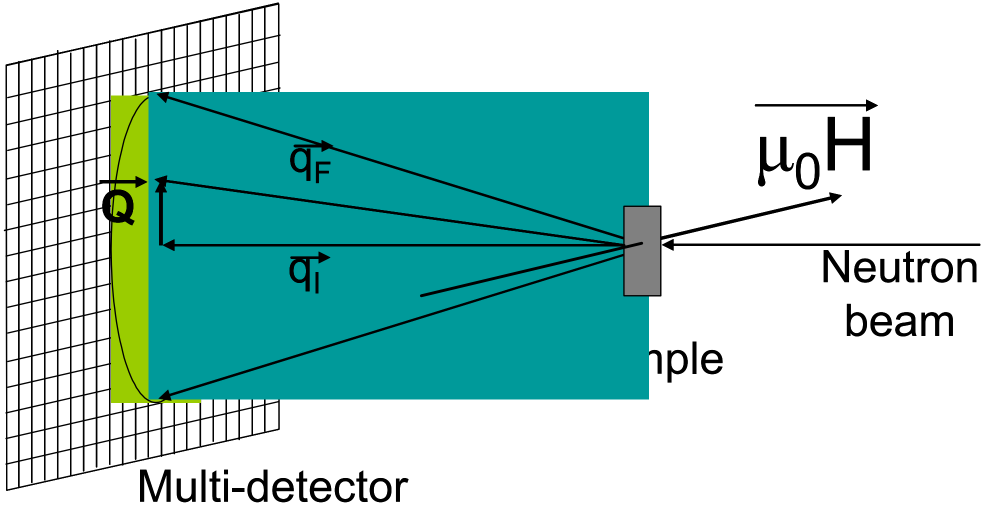
Classical 3D percolation models give : 1.5<s<2 et $\Phi_T$ ~ 15%

***V. Hardy et al. , Phys. Rev. B 64, 64402 (2001) :*** $S = 3.6 \quad \phi_T \approx 4\%$

⇨ Due to correlations between nanometric clusters and/or anisotropic geometry of these later ?

# THE SMALL ANGLE NEUTRON SCATTERING TECHNIQUE (SANS) :





Measure of the correlated disorder

$$I(\vec{Q}) = \int_V \tilde{\rho}^2(\vec{r})\exp(i\vec{Q}\cdot\vec{r})d\vec{r}$$

Observation of the magnetic inhomogeneities in the range 2-200 nm

*J. M. De Teresa et al. , Nature 386, 256 (1997)*

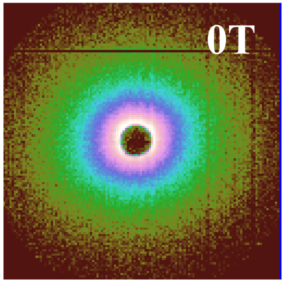
# SCATTERED INTENSITY :

Total intensity : $I = I_N + I_M$ Single crystals : $I_N << I_M$

Magnetic intensity : $I_M(Q,H) = V_p P(Q)\phi(1-\phi)\left\langle(\Delta\rho_M(H))^2\right\rangle$

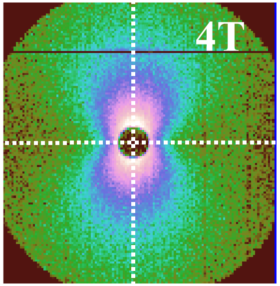
Density of scattering length : $\rho_i = \delta m_i \sin\alpha_i$ $\alpha_i = \left(\vec{Q},\vec{m}_i\right)$

$\mu_0$H=0



$$\left\langle(\Delta\rho_M(H))^2\right\rangle$$
$$= \delta^2 m_F{}^2 \left\langle \sin^2\alpha_F \right\rangle$$
$$= \delta^2 m_F{}^2 \frac{3}{2}$$

$\mu_0$H>1T horizontal



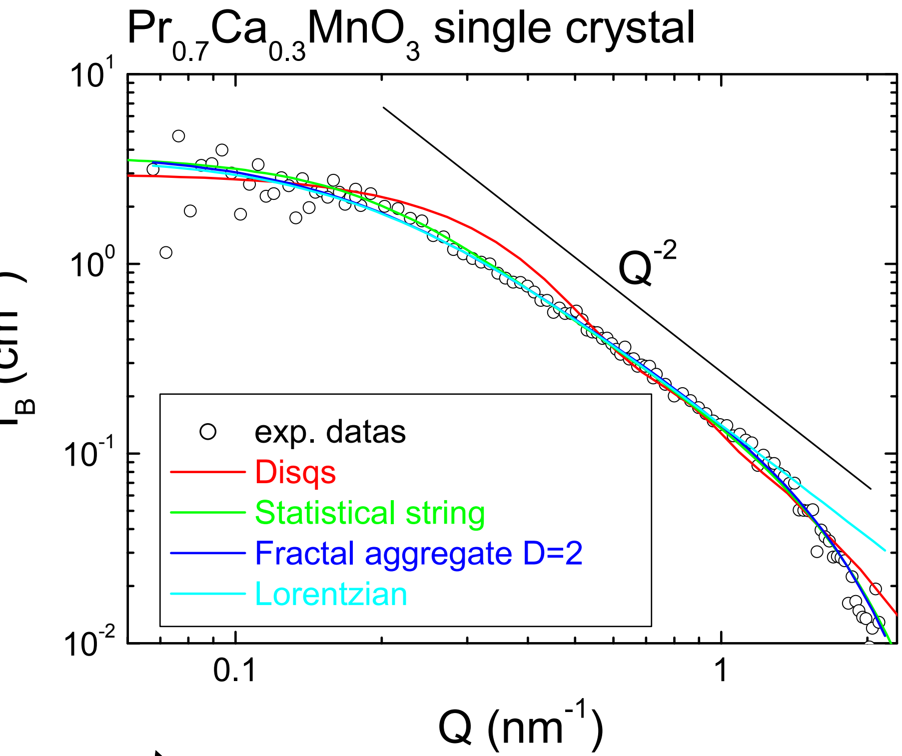
$$\left\langle(\Delta\rho_M(H))^2\right\rangle_O$$
$$= \rho^2\left(m_F - \chi_{AF}\mu_0 H\right)^2 \sin^2\alpha$$
$$\sin^2\alpha = \sin^2\left(\vec{Q},\vec{H}\right)$$

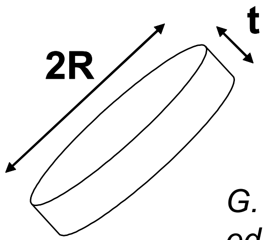
$$I(Q,\alpha) = I_A(Q) + I_B(Q)\sin^2\alpha$$

$I_A$: non oriented magnetism and nuclear signal

$I_B$: oriented magnetism

# SANS AT 2K UNDER 2T
# X=0.3 SINGLE CRYSTAL :



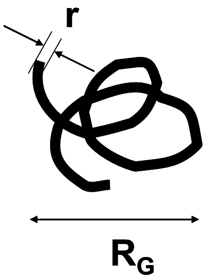


Disks :

**R ~ 10 nm t ~ 1.1 nm Φ ~ 5%**

*G. Porod, in Small Angle X-ray Scattering, edited by O. Glatter and O. Kratky, Academic Press, London, 1983, p. 35*

*Ch. Simon et al. , Phys. Rev L 89, 207202 (2002*

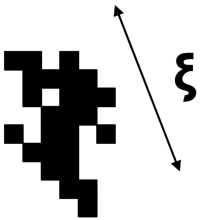


Statistical strings :

**$R_G$ ~ 7.5 nm r ~ 1.3 nm Φ ~ 3%**

*J. S. Pedersen et al. , Macromolecules **29**, 7602 (1996)*

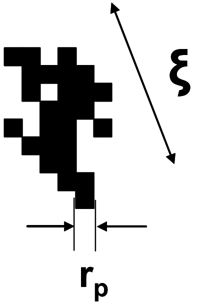
*M. Viret et al. , Phys. Rev. Lett. 93, 217402 (200*



Lorentzian :

**ξ ~ 5 nm**

*J. M. De Teresa et al. , Nature 386, 256 (1997)*

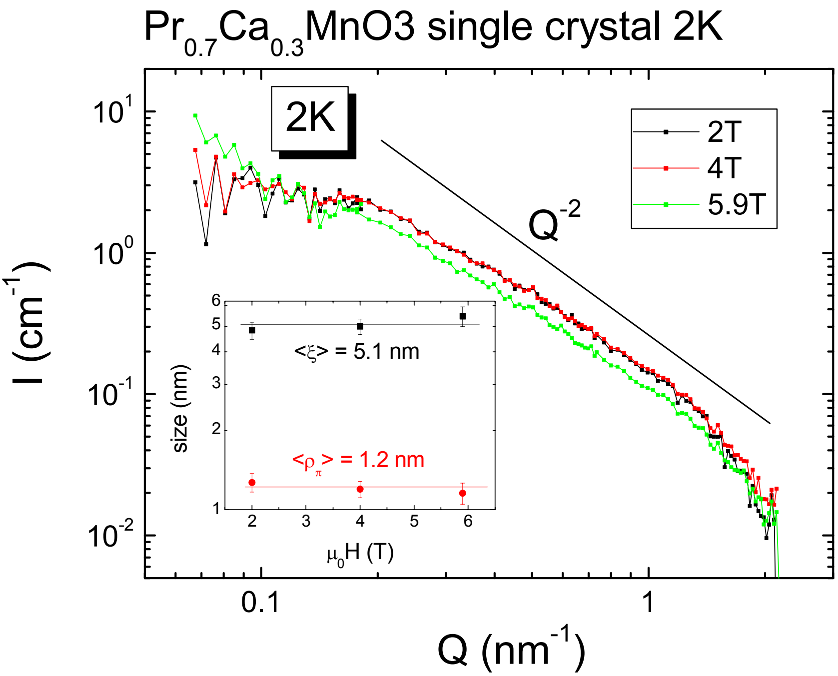


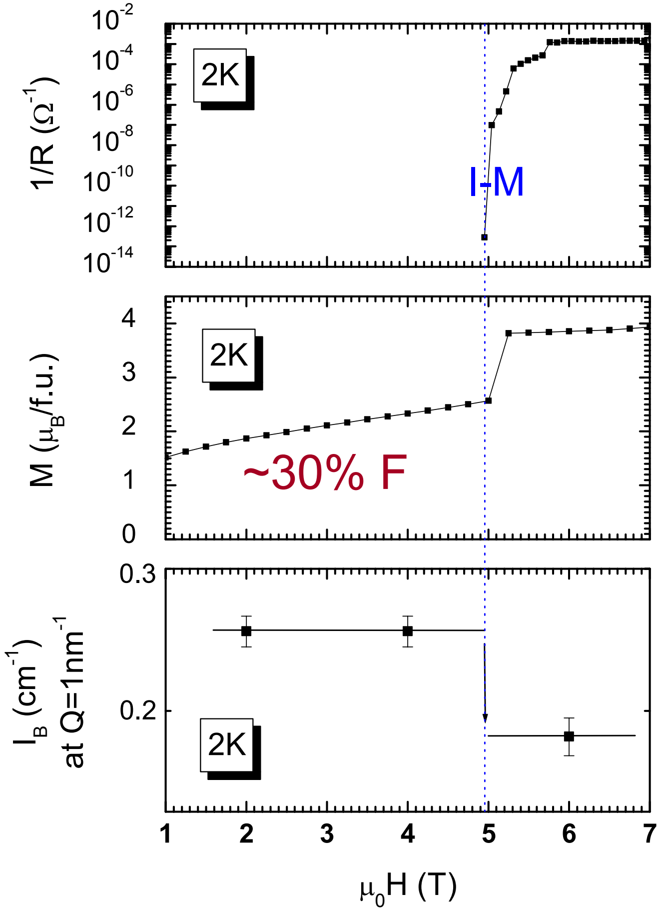
Fractal Aggregate D=2 :

**ξ ~ 5.3 nm $r_p$ ~ 1.7 nm Φ ~ 2%**

*J. Teixeira, J. Appl. Phys. **21**, 781 (1978)*

# SANS AT 2K
# X=0.3 SINGLE CRYSTAL :

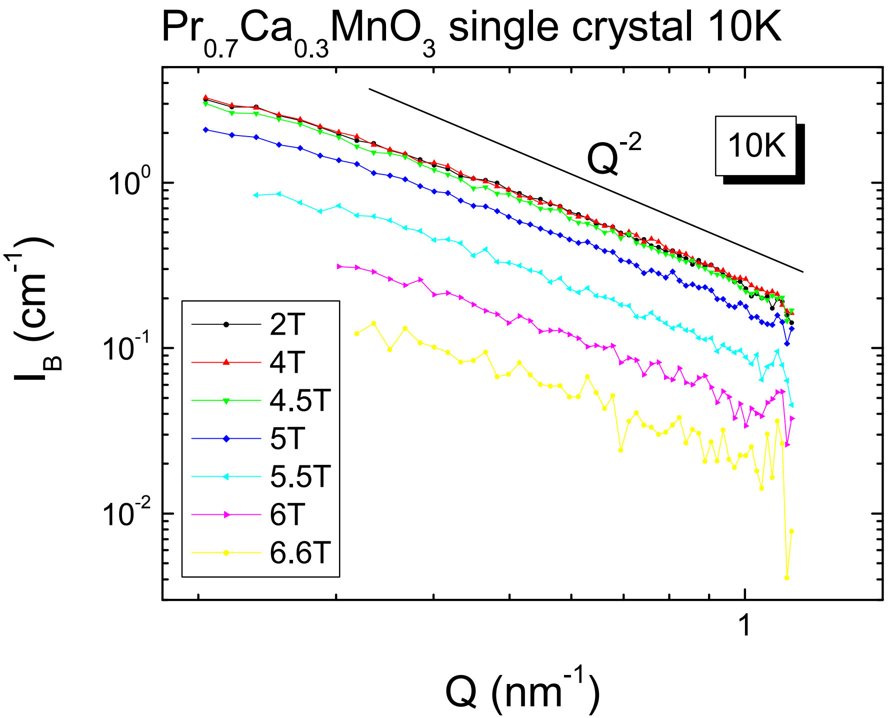


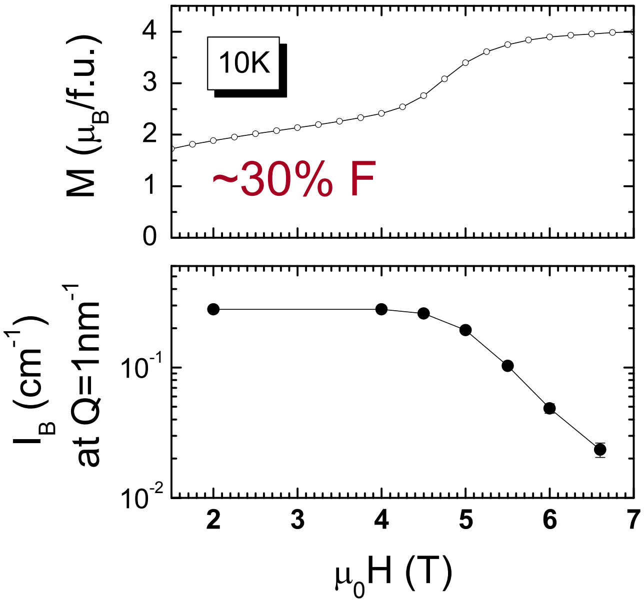


- A 1rst order metamagnetic transition
- A simultaneous 1rst order I-M transition
- The size and shape of clusters never change, only the magnetic contrast change

**The I-M transition is not a classical percolation phenomena**

# SANS AT 10K and 30K
# X=0.3 SINGLE CRYSTAL :


$Pr_{0.7}Ca_{0.3}MnO_3$ single crystal 10K

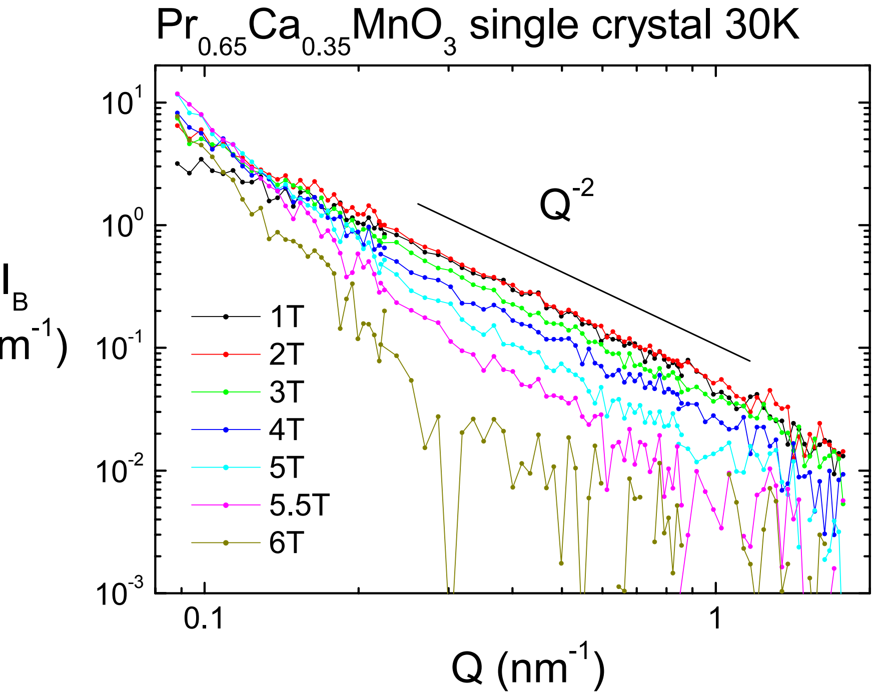


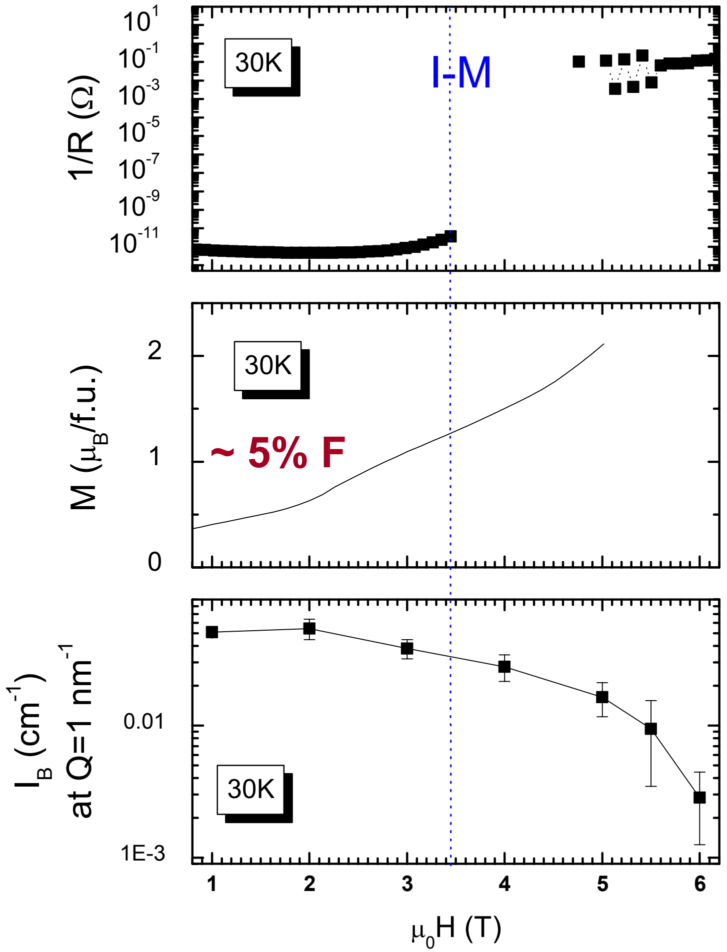
**No change of size and shape of the F clusters**

**A second order metamagnetic transition characterized by a loss of magnetic contrast and ot a nucleation and growth of F clusters**

**he I-M transition is still not a classical percolation phenomena**

# SANS AT 30K
# X=0.35 SINGLE CRYSTAL :



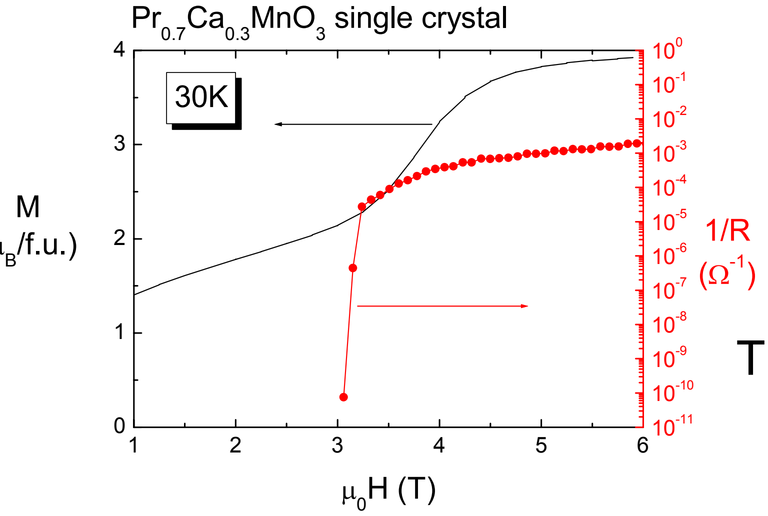


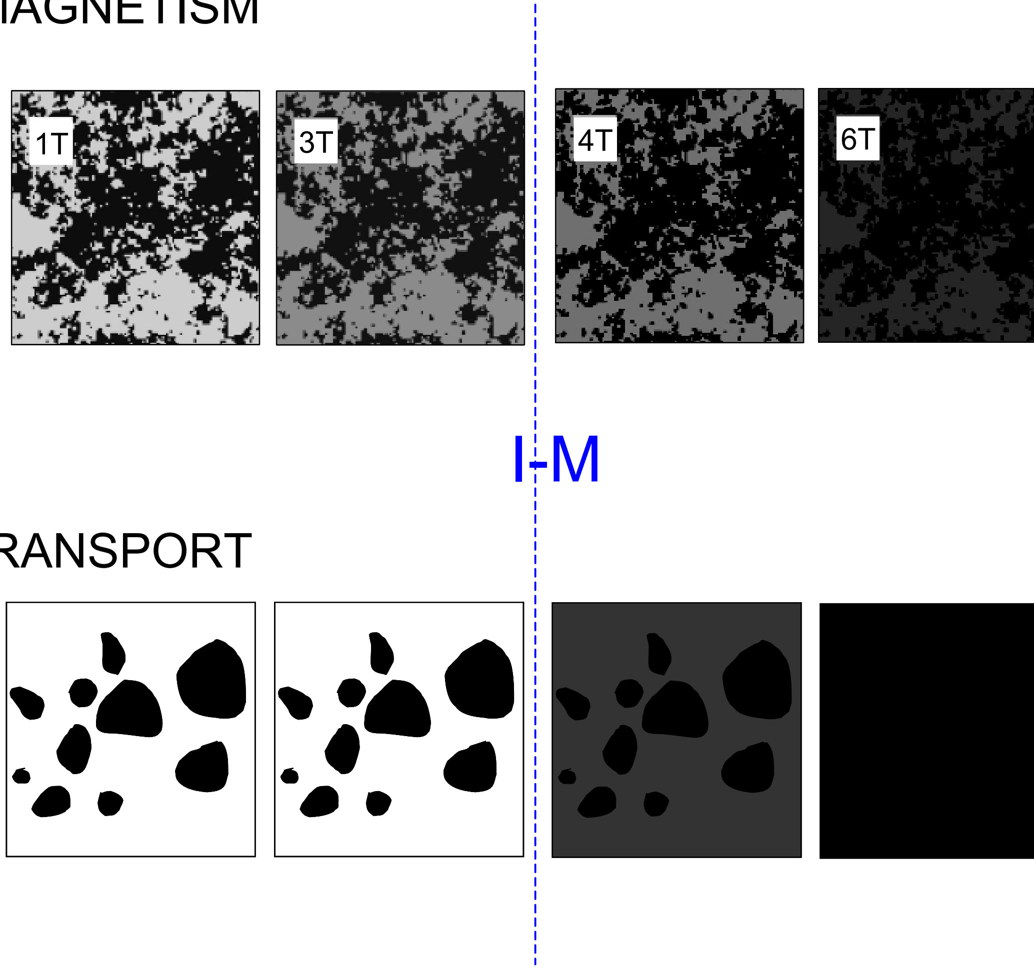
No change of size and shape of clusters

A 2nd order metamagnetic transition :

loss of magnetic contrast and not nucleation or growth of F clusters

# SANS AT 30K
# 0.3<X<0.35 SINGLE CRYSTALS :



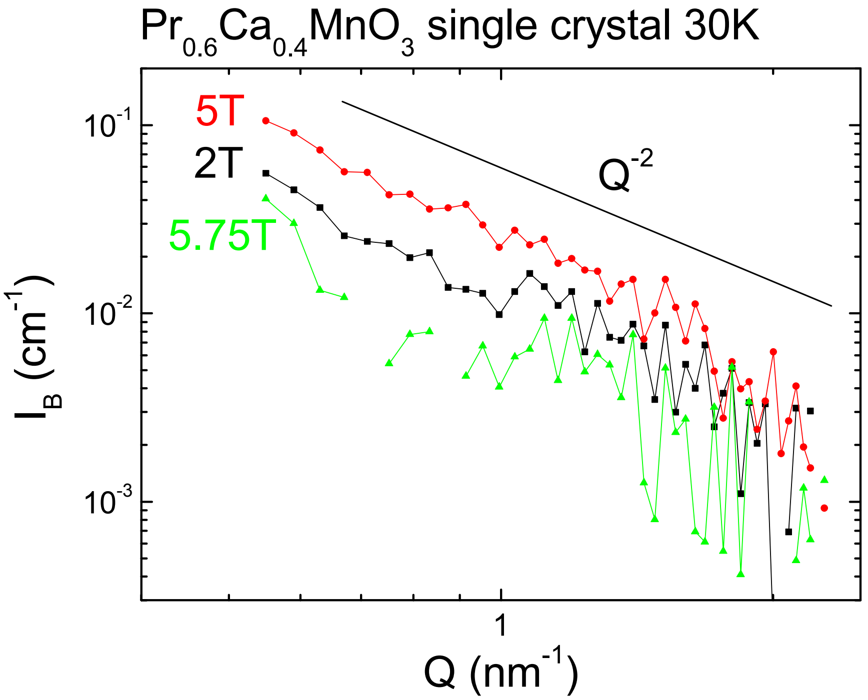


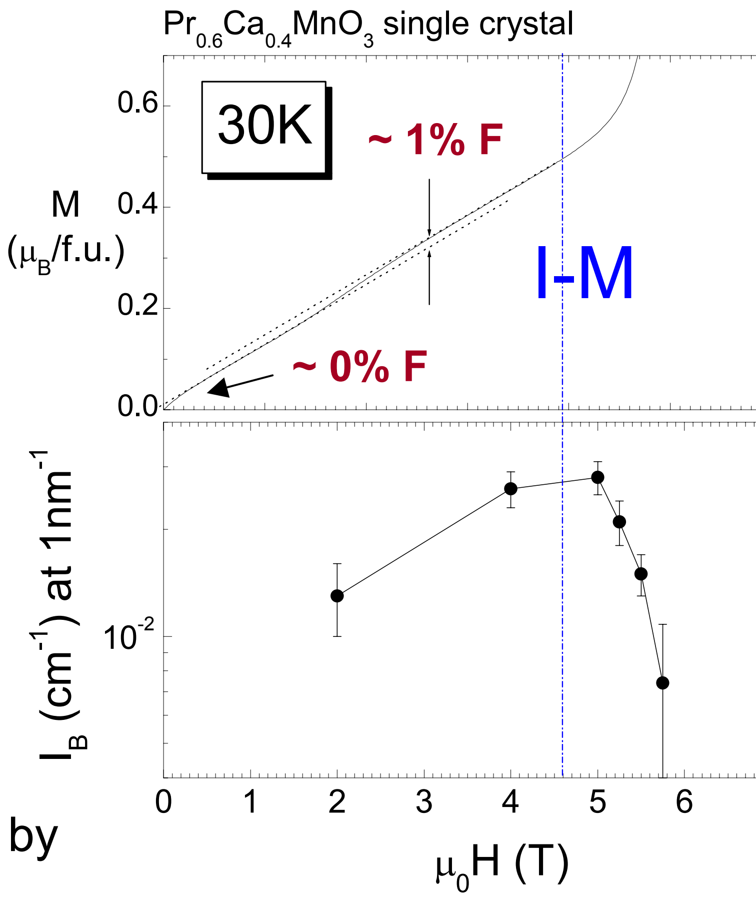
MAGNETISM

I-M

TRANSPORT

# SANS AT 30K
# X=0.4 SINGLE CRYSTAL :





**NUCLEATION OF CLUSTERS**

A 2nd order metamagnetic transition characterized by loss of magnetic contrast

# CONCLUSIONS :

SANS is a powerful technique to study the nanometric magnetic phase separation

CMR compounds $0.3<x<0.4$ show nanometric F clusters in the Af matrix under field

HE AF-to-F METAMAGNETIC TRANSITIONS FOR $0.3<X<0.4$ :

It corresponds to a loss of magnetic contrast

1rst order at 2K

2nd order at 30K

RANSITION I-M FOR $0.3<X<0.35$ :

1rst order: evolution by steps

Complex behavior which differs from a classical percolation scenario :

- percolative quantum transport? (*S. Kumar et al. , Phys. Rev. Lett. 92, 126602*)
- avalanche phenomena due to complex properties of clusters interfaces?

(*J. Burgy et al. , Phys. Rev. B 67, 014410 (2003)*)

# CONCLUSIONS :

## M TRANSITION IN X=0.4 SINGLE CRYSTAL :

Sharp at the critical field, progressive at higher fields

Nucleation of clusters under magnetic field :

- Fits well with classical percolation models with low critical phase fraction and high percolation exponent

*V. Hardy et al. , Phys. Rev. B 64, 064402*