

Glassy charge dynamics and magnetotransport in lightly doped La₂CuO₄

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- High-T_C superconductors: doped Mott insulators
- Metal-insulator transition; Coulomb glass
- Role of inhomogeneities in cuprates one of the major open issues
- Nature of the ground state in cuprates? Case of La₂CuO₄ at low doping:
 - hysteretic and memory effects in magnetotransport
 - resistance fluctuations (noise)
 - microscopic mechanism
- Conclusions and future work

Complex behavior of high-T_C superconductors



- **3D Néel order at 310 K** in the parent material
- 3D Néel order suppressed with ≈2-3% doping
- only a few per cent
 (≈5-6%) of dopants cause
 a transition from an
 insulating to a (super)
 conducting state
- undoped parent compound (e.g. La₂CuO₄): Mott insulator, not a metal
- single-electron band theory of solids fails

Parent cuprate La₂CuO₄





- one hole on each Cu site
- in-plane AF exchange interaction $J_{\parallel} \sim 0.1~eV$
- weak inter-plane interaction $(J_{\parallel}/J_{\perp} \sim 10^5)$

3D long-range antiferromagnetic (AF) order:



• weak FM moments along c-axis in each ab plane due to spin canting



How does a transition from a Mott insulator to a conductor occur as a function of doping?

General problem

- high density kinetic energy (Fermi energy) dominates
- low density potential energy dominates:
- electron-electron interactions(Mott insulator)

disorder due to impurities, defects (Anderson insulator)



Result: formation of localized (bound) states \longrightarrow no conduction





•"dynamical scaling" in the critical region: $\sigma(n_s,T) \propto T^x f(T/\delta_n^{zv})$

• power-law critical behavior: $\sigma(n_s, T=0) \propto \delta_n^{\mu}$

Theoretical problems: no broken symmetry; order parameter? No small parameter; elementary excitations? Standard approaches fail

It gets even more complicated...



- Coulomb repulsion: keep electrons apart (uniform density)
- Random potential: nonuniform density
- competition between Coulomb interactions and disorder



Coulomb glass



 expected in Anderson insulators with strong electron-electron interactions [M. Pollak (1970); Efros, Shklovskii (1975); Davies, Lee, Rice (1982,84)]

Observations of glassiness in electronic systems – very few:

- slow relaxations in GaAs capacitance (Monroe et al.)
- slow relaxations and thermal hysteresis in conductivity of granular films (Goldman *et al.*, Wu *et al.*, Frydman *et al.*)
- slow relaxations of photoconductivity in $YH_{3-\delta}$ (Lee *et al.*)
- slow relaxations, aging, memory in conductivity of InO_x (<u>Ovadyahu *et al.*</u>) and granular Al (Grenet *et al.*)
- 2D electrons in Si (DP *et al.*): slow relaxations, aging, memory; slow, correlated dynamics – from insulating to (poorly) metallic

my work

• lightly doped cuprates (DP et al.)







3D long-range antiferromagnetic (AF) order:



- weak FM moments along c-axis in each ab plane due to spin canting
- doped holes go into the 2D CuO₂ planes
 - ⇒ Long-range AF order destroyed for x ≥ 0.02 (Sr), x>0.03 (Li) short-range magnetic order persists



• AF domains; holes along (antiphase) domain walls?



carrier concentration

• spin glass behavior well established at low T

Nanoscale charge inhomogeneities



$Ca_{2-x}Na_{x}CuO_{2}Cl_{2}$

[Kohsaka *et al.*, Phys. Rev. Lett. 93, 097004 (2004)]

• global phase separation not possible because of <u>charge neutrality</u>





("stripe- and clumpforming systems")

[Reichhardt *et al.*, Europhys. Lett. 72, 444 (2005)]





Doped Mott insulatorsInsulator to (super) conductor transition?





Glassy charge dynamics in cuprates? Nature of the ground state?

<u>Glasses:</u> - many metastable states - slow, nonequilibrium dynamics

How to probe glassy dynamics?

supercooled water

- measure response of the system to some kind of a perturbation ⇒ slow, nonexponential relaxations
- fluctuations provide complementary information (correlations)

For charge glass: transport (bulk probe; mean values of resistivity) and resistance noise (fluctuations)





- a good candidate (well characterized, disorder)

• Sr and Li (no magnetic moment) doped: similar magnetic behavior, but no SC in Li-LCO x=0.03 in this talk: **Femperature** Sr²⁺ $La_2Cu_xLi_{1-x}O_4$ in antiferromagnetic С AF phase ($T_f \sim 7-8$ K) O Cu²⁺ SG $La_{2-x}Sr_{x}CuO_{4}$ in O²⁻ Х Li-LCO LSCO spin-glass phase 🔘 La³⁺ $(T_{sg} \sim 7-8 \text{ K})$

Lightly doped La₂CuO₄

Dielectric measurements: an electronic glass state AF Li-LCO: Park et al., PRL 94, 017002 (2005); x=0.03 LSCO: Jelbert et al., PRB 78, 132513 (2008)







а

Li¹⁺

Temperature dependence of the resistance



Variable-range hopping (VRH): $R \propto \exp[(T_0/T)^n]$, n=1/3 (2D exponent) Actually, it depends on how the sample is cooled...

History dependence in transport: zero-field cooling (ZFC) vs. field cooling (FC)



Difference between FC and ZFC resistance R(B=0)





- higher B enable overcoming higher energy barriers
- R(B=0) determined by the highest B previously applied - memory of magnetic history

Also, manganites: Levy et al., PRL 89, 137001 (2002); YBCO: Ando et al., PRL 83, 2813 (1999)

Hysteretic behavior of the resistance

Memory in R wiped out for T \ge 1K, spin glass transition T_{SG} \approx 7K

 $(\mathbf{R}_{c}, \mathbf{R}_{ab};$

B||c and **B**||ab)



- return point memory
- incongruent subloops ⇒ interactions between domains

Resistance fluctuations (noise); LSCO





- noise Gaussian at "high" T
 (e.g. T > 0.18 K for R_{ab} noise)
- at low T, non-Gaussian noise
 - metastable states (out-of-equilibrium)



Probability density functions (PDF) of fluctuations



 structure depends on the observation time – different states contribute ⇒ nonergodic





- increase sampling time to 12 hours, but never becomes
 Gaussian at low T
- nonergodic, does not reach
 equilibrium on experimental
 time scales at low T

Onset of glassiness in transport at T<< T_{SG}: suggests spin and charge glass not directly related





T (K)



8

0.2

I || ab

6

Noise statistics independent of

both **B** and magnetic history



- Noise statistics independent of both B and magnetic history (unlike conventional spin glasses) ⇒ charge, not spin!
- Onset of hysteretic and memory effects in magnetoresistance: $T_{onset} \ll T_{sg}$

 \Rightarrow Charge glass transition $T_{cg}=0$

[I. Raičević et al., PRL101, 177004 (2008)]

Scaling of the second spectra



- can distinguish between different models:
- droplet approach
- hierarchical, tree-like model

S₂ decreases with f for a fixed f₂/f, consistent with droplet picture (short-range interactions)

Spatial segregation of holes as a result of competing interactions on different length scales

Cluster charge glass





Origin of large positive magnetoresistance at low T???





Out-of-plane MR - LSCO

In-plane MR - LSCO

<u>High T</u> – MR negligible

- negative below 10K (onset of spin glass order)
- isotropic
- reorientation of weak FM moments

<u>Low T</u> Emergence of low-field positive MR at T < 1K

Strong positive MR at low T (below 0.5 K)

Low-T positive MR coincides with the onset of charge glassiness **Positive MR - glassy features:**

Only positive MR exhibits

hysteresis

83

∆R_{ab}/R_{ab}

2

B (T)

6

T = 0.464 K

6

8

4

B (T)

8

• History dependence

2

T = 0.120 K

B || ab

LSCO

• Memory

20

15

10

5

0

- Hysteresis
- In the same regime:
- \bullet Noise – glassy dynamics as $T \rightarrow 0$

Out-of-plane MR – Li-LCO

[Similar to AF (x=0.01) LSCO: Ando *et al.*, PRL 90, 247003 (2003)]

<u>High T</u>

- B || c
 - negative steplike decrease (spin flop)
 - strong positive below $\sim 12~K$
- B || ab
 - negative (∝ B²; smooth rotation of weak FM moments)

Low T

• B || ab: low-field positive MR

below 3 K

5

0

-5

-10

-15

2

In-plane MR – Li-LCO

History dependent behavior

R_{FC} – R_{ZFC} decreases with increasing T and vanishes at a B-dependent T

- Memory the highest applied field determines R(B=0)
- Hysteresis only in the region of (initial) positive MR
- Return point memory

Must be a spin related effect!!

Reorientation of weak ferromagnetic moments leads to

Negative, not positive MR

<u>Remaining possibility: coupling of B to the spins of holes</u> which populate localized states within Mott-Hubbard gap U Strongly disordered materials with Mott VRH and intra-state correlations (Coulomb repulsion U' between two holes in the same disorder-localized state)

Positive MR in various nonmagnetic, disordered materials with strong Coulomb interactions attributed to this effect

Meir, Europhys. Lett. 33, 471 (1996): Positive MR due to Zeeman splitting - universal function of B/T log R

• It appears that the magnetic background remains inactive in this regime of T and B (frozen spins/AF domains; holes that "live" in domain walls – analogous to other disordered, interacting systems)

Conclusions

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Lightly doped La₂CuO₄ : two different transport regimes within the spin glass phase

1) <u>"High T"</u> < T_{sg}: - magnetic structure important \Rightarrow negative MR

- **2)** <u>Low T,</u> $T \rightarrow 0$ limit (*i.e.* T < 1 K in practice):
 - glassy charge dynamics (noise); charge cluster glass, T_{cg}=0
 - positive MR with hysteresis and memory
 - magnetic structure not important, to leading order
 - U' on disorder-localized state important (U'~20 K in LSCO)

As T→0, behavior characteristic of systems that are far from any magnetic ordering

Use hysteretic, **positive MR** as an **easy tool** for detecting charge glassiness confined to the domain walls: intrinsic or driven by disorder?

Поповић, Београд '09