The room temperature precursor of the low-temperature ordered phases in copper oxides

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i-Lamp
(Interdisciplinary laboratories for advanced materials physics)
• Spontaneous symmetry-breaking in copper oxides: charge order

• Ultrafast spectroscopies to investigate the role of the Mott-like excitations

• The room temperature precursor of charge-order in cuprates
People and Collaborations

• **Ultrafast optics group** (Università Cattolica, Brescia)
  S. Peli, N. Nembrini, F. Banfi, G. Ferrini, C. Giannetti

• **Ultrafast optics group** (Università degli Studi di Trieste)
  F. Cilento, D. Fausti, F. Parmigiani

• **Ultrafast optics group** (Politecnico di Milano)
  S. Dal Conte, D. Brida, G. Cerullo

• **Equilibrium spectroscopies**
  R. Comin, B. Ludbrook, A. Damascelli (University of British Columbia, Vancouver)
  M. Greven (University of Minnesota & Stanford University)
  B. Keimer (MPG-UBC center for QM)

• **Non-equilibrium models of correlated materials**
  L. Vidmar (LMU Munich), M. Mierzejewski (Katowice), J. Bonca (Ljubljana)
  M. Capone, M. Fabrizio (SISSA, Trieste)

• **Equilibrium optical properties of HTSC**
  D. Van der Marel (Université de Genève)
  S. Lupi (La Sapienza, Roma)
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The phase diagram of copper oxides

Superconductivity on top of a very unconventional ground state!
charge-order

phase diagram

temperature $T$

$T_{cdw}$

$T^*$

$p$


ARPES underdoped $T_c=15K$

X-ray scattering

STM on Bi2212


$Q_{HS}=0.255$

$Q_{AF}=(\pi,\pi)$

($\pi,\pi$)

($\pi,0$)

hot spots

Bi2201
charge-order

open problems:

• interplay with superconductivity
• origin of charge-order
• relation with the “Mott” physics

STM on Bi2212

R. Comin et al. Science 343, 390 (2014)

Relation between charge-order and the Mott physics

Is there any relation between the low-temperature charge-order and the Mott physics?

“Mott” physics in copper oxides

charge-transfer process at $\Delta_{CT} \approx 2$ eV

need for a high-energy probe $\gg K_B T$

Cu $3d^9 \rightarrow$ Cu $3d^{10} + h_0$
Outline

• Spontaneous symmetry-breaking in copper oxides: charge order

• Ultrafast spectroscopies to investigate the role of the Mott-like excitations

• The room temperature precursor of charge-order in cuprates

• Snapshots of the retarded-interaction with ultrafast fluctuations via 10 fs pulses
  S. Dal Conte et al., Science 335, 1600 (2012)
the energy scales in optics (copper oxides)

equilibrium optical conductivity

\[ \sigma(\omega) \]

\[ \gamma(\omega, T) \]

\[ U > 0 \]

\[ \Delta_{CT} \]

2J

energy
equilibrium optical conductivity

\[ \sigma(\omega) \]

\[ \gamma(\omega, T) \]

energy

\[ U > 0 \]

\[ \Delta_{CT} \]

charge-transfer process

at \( \Delta_{CT} = 2 \text{ eV} \)

\[ \text{Cu } 3d^9 \rightarrow \text{Cu } 3d^{10} + h\omega \]

probe of the local correlations
the energy scales in optics (copper oxides)

equilibrium optical conductivity

\[ \gamma(\omega, T) \]

\[ \sigma(\omega) \]

\[ U > 0 \]

\[ \Delta_{CT} \]

\[ 2J \]

charge-transfer process at \( \Delta_{CT} \approx 2 \text{ eV} \)

\[ \text{Cu } 3d^9 \rightarrow \text{Cu } 3d^{10} + h_0 \]

strongly-correlated scenario:

probe of local correlations through the CT process

white-light probe

pump

\[ h_0 \]

\[ \text{O } 2p \]

probe of the local correlations
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Equilibrium optical properties of Bi2201

$\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$

$\gamma(\omega, T)$

$\sigma(\omega)$

$U > 0$

$\Delta_{CT}$

energy
Equilibrium optical properties of Bi2201

\[ \text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_6+\delta \]

\[ p=0.10, \ T_c=0 \]

\[ p=0.18, \ T_c=19 \text{ K} \]
A non-equilibrium phase diagram for cuprates

UNDERDOPED SYSTEMS: $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$

$T=300\ \text{K}$

$p=0.10,\ T_c=0$
A non-equilibrium phase diagram for cuprates

UNDERDOPED SYSTEMS: $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$

- $p=0.12$, $T_c=13$ K
- $p=0.10$, $T_c=0$
A non-equilibrium phase diagram for cuprates

UNDERDOPED SYSTEMS: $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$

- $p=0.13$, $T_c=17$ K
- $p=0.12$, $T_c=13$ K
- $p=0.10$, $T_c=0$

Doping vs. temperature diagram
A non-equilibrium phase diagram for cuprates

UNDERDOPED SYSTEMS: Bi$_2$Sr$_{2−x}$La$_x$CuO$_{6+δ}$

- $p=0.12$, $T_c=13$ K
- $p=0.13$, $T_c=17$ K
- $p=0.16$, $T_c=33$ K
- $p=0.10$, $T_c=0$
A non-equilibrium phase diagram for cuprates

UNDERDOPED SYSTEMS: Bi$_{2}$Sr$_{2−x}$La$_{x}$CuO$_{6+δ}$

$p=0.12$, $T_c=13$ K

$p=0.13$, $T_c=17$ K

$p=0.16$, $T_c=33$ K

$p=0.10$, $T_c=0$

$p=0.18$, $T_c=19$ K
Main facts

\( \text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta} \)
\( p=0.12, T_c=13 \text{ K} \)

- transient redshift of the CT peak
- independent of the pump energy (1.5, 1.9, 3 eV)
CT redshift

$\text{Cu } 3d^9 \rightarrow \text{Cu } 3d^{10} + h\nu$

$U_{pd}$

O-2$p_x$

Cu-3$d_{x^2-y^2}$

O-2$p_y$
CT redshift

Cu $3d^9 \rightarrow Cu \ 3d^{10} + h\Omega$

$U_{pd}$

O-2$p_x$

O-2$p_y$

Cu-3$d_{x^2-y^2}$

Local picture: localized excitons with binding energy $\propto U_{pd}$

$$\delta \Delta_{CT} = - \left( 2U_{pd} - \frac{5}{24} U_{pp} \right) |\delta \epsilon_\downarrow|$$

mean-field calculation by M. Fabrizio

see also B. Mansart et al. *PNAS* **110**, 4539 (2013)
A non-equilibrium phase diagram for cuprates

$p_{cr}=0.16 \pm 0.01$

- transition in the high-energy physics (CT redshift) at $T=300K$
A non-equilibrium phase diagram for cuprates

\[ p_{cr} = 0.16 \pm 0.01 \]

- transition in the high-energy physics (CT redshift) at \( T = 300K \)
- disappearance of the charge-order

CDW measured by RXS

R. Comin, Science 343, 390 (2014)
Conclusions

low-temperature CDW emerges only for $p < p_{cr}$

→ consequence of a precursive correlated state
Conclusions

• charge-order is the low-energy manifestation of a correlated state dominated by the local Cu-3d–O-2p interactions

• the quenching of the O-2p→Cu-3d charge fluctuations at the energy scale Δ_{CT} plays a relevant role

• the oxygen orbitals are strongly involved→beyond single-band Hubbard model

• Does the high-energy transition at p≈0.16 have more general consequences?
Conclusions

- charge-order is the low-energy manifestation of a correlated state dominated by the local Cu-3d–O-2p interactions

- the quenching of the O-2p→Cu-3d charge fluctuations at the energy scale $\Delta_{\text{CT}}$ plays a relevant role

- the oxygen orbitals are strongly involved→beyond single-band Hubbard model

- Does the high-energy transition at $p \approx 0.16$ have more general consequences?

$p_{\text{cr}} \rightarrow$ transition from closed Fermi surface to disconnected arcs

Y. He et al. Science 344, 608 (2014)
Conclusions

• charge-order is the low-energy manifestation of a correlated state dominated by the local Cu-3d–O-2p interactions

• the quenching of the O-2p→Cu-3d charge fluctuations at the energy scale $\Delta_{CT}$ plays a relevant role

• the oxygen orbitals are strongly involved→beyond single-band Hubbard model

• Does the high-energy transition at $p \approx 0.16$ have more general consequences?

$\rho_{cr} \rightarrow$ transition from kinetic energy gain to k.e. loss

TRENDOXIDES2015

School and Workshop: New TRENDS in Correlated OXIDES and Interfaces

12-18 November, Brescia

• Oxides: cuprates, vanadates, manganites and other
  3d-4d compounds
• Oxide interfaces and superlattices
• Correlated-oxide based devices
• Non-equilibrium phenomena in oxides
• Models and theory for materials and spectroscopies
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we are waiting for you in Brescia!!!
Thank you!
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Extended Drude Model

equilibrium optical conductivity

\[ \sigma(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + M(\omega, T))} \]

Extended Drude dielectric function

\[ \epsilon_D(\omega, T) = 1 - \frac{\omega_p^2}{\omega(\omega + M(\omega, T))} \]

Memory function
ultra high temporal resolution

-50 0 50

delay (fs)

fwhm: \(18\pm2\) fs

non-linear crystal

\(\omega_1\pm\omega_2\)

G. Cerullo’s group (Politecnico of Milan)

optimally doped \(\text{Bi}_2\text{Sr}_2\text{Y}_{0.08}\text{Ca}_{0.92}\text{Cu}_2\text{O}_{8+\delta}\) (YBi2212)

\(T_c=96\) K
In 16 fs photoexcited carriers can exchange energy with bosons

\[ \delta R(\omega) \propto \delta T_b \]

\[ \approx 40 \text{ fs} \Rightarrow \tau_r = 16 \pm 3 \text{ fs} \]

Summary

High $T_c$ superconductor (YBi2212)
$T_c=83$ K

BCS superconductor ($MgB_2$)
$T_c=39$ K

Metal (Gold)
$T_c=0$ K

Spin fluctuations
Strongly coupled phonons
Lattice

$\tau_r = 16 \pm 3$ fs

Hot electrons
$E_F$

$\tau_r = 31 \pm 3$ fs

Delay time

$\tau_r \sim 600$ fs

Critical Temperature

Critical Temperature

$T_c$(K)

Summary

100
80
60
40
20
0
-20
-40
-60
-80
-100
-120

Critical Temperature
electron-boson coupling in CUPRATES

\[ \Pi(\Omega) = \alpha^2 F(\Omega) + I^2 \chi(\Omega) \]


In agreement with the glue extracted from the Hubbard model

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