30 years of high Tc:
Superfluid and normal-fluid densities in the cuprate superconductors

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Possible High $T_c$ Superconductivity in the Ba–La–Cu–O System

J.G. Bednorz and K.A. Müller
IBM Zürich Research Laboratory,
Received April 17, 1986

Fig. 1. Temperature dependence of resistivity in Ba$_2$La$_2$–Cu$_2$O$_{4+δ}$. 
April 17, 1986 – just over 30 years!

2 Get Nobel for Unlocking Superconductor Secret

By James Gleick

With a swirl that echoed the rush of discovery over the last few months, the Nobel Prize in Physics was awarded yesterday to two scientists in Switzerland: whose breakthrough just last year has touched off a torrent of research in the long-dormant field of superconductivity.

The scientists, J. Georg Bednorz and K. Alex Müller, after learning they had won the Nobel Prize in Physics.

Scientists said in its announcement from Stockholm yesterday, “This set off an avalanche.”

Physicists have already surpassed the discovery with new superconductors that become superconducting at higher temperatures, raising the prospect of applications from high-temperature superconductors in new ways, from small generators and power grids to huge supercomputers and nuclear fusion devices.

They are J. Georg Bednorz, left, and K. Alex Müller after learning they had won the Nobel Prize in physics.
• 1911: H. K. Onnes, who had figured out how to make liquid helium, used it to cool mercury to 4.2 K and looked at its resistance:
  
  - Current can flow, even if $E=0$.
  - Current in superconducting rings can flow for years with no decrease!

• 1933: Meissner effect: Magnetic field is zero inside a superconductor!

  
  - Bardeen’s second Nobel prize (1956 – transistor)

• 1986: Bednorz and Mueller discover HTSC.
  
  - No longer a low temperature phenomenon
The Meissner Effect

- A diamagnetic property exhibited by superconductors.

- End result is the exclusion of magnetic field from the interior of a superconductor.
Materials

- **YBCO == 123**: $\text{YBa}_2\text{Cu}_3\text{O}_7$ 94 K
- **BSCO == 2212**: $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ 91 K
- **LSCO == 214**: $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ 39 K
Materials

- YBCO == 123: \( YBa_2Cu_3O_7 \)
- BSCO == 2212: \( Bi_2Sr_2CaCu_2O_8 \)
- LSCO == 214: \( La_{2-x}Sr_xCuO_4 \)
- TBACO up to 110 K
- HgBaCuO up to 134 K (153 K under pressure)
Superconductivity timeline

Critical temperature $T_c$ [K]

Year


- liq. CF$_4$
- liq. N$_2$
- liq. H$_2$
- liq. He

Materials and Phases:
- HgBaCaCuO @ 30 GPa
- HgTlBaCaCuO
- Cs$_3$C$_60$ @ 1.4 GPa
- MgB$_2$
- SrFeAs
- FeSe
- H$_2$S @ 155 GPa

Compounds:
- BiSrCaCuO
- TlBaCaCuO
- YBaCuO
- LaSrCuO
- BaCuO
- BKBO
- YbPd$_2$B$_2$C
- RbCsC$_60$
- K$_3$C$_60$
- Li @ 33 GPa
- PuCoGa$_5$
- PuRhGa$_5$
- CeCoIn$_5$
- CNT
- diamond
- LaOF$_3$FeP$_3$
What sort of materials are the cuprates?

- La$_2$CuO$_4$ is the “parent” material
- Would be half filled band metal
What sort of materials are the cuprates?

- $\text{La}_2\text{CuO}_4$ is the “parent” material
- Would be half filled band metal
- Layered, with square-planar $\text{CuO}_2$ sheets
What sort of materials are the cuprates?

- La$_2$CuO$_4$ is the “parent” material
- Would be half filled band metal
- Layered, with square-planar CuO$_2$ sheets
- Actually a “charge-transfer” insulator
Phase diagram

- Charge carriers are holes.
- Adding holes (above ~5%) produces superconductivity.
- “Optimal doping”
  - $\Rightarrow$ max $T_c$
  - $\Rightarrow$ linear resistivity
  - “strange metal”
Charge transport

- **Cuprates:**
  “metallic” dc resistance

- \( \rho = A + B T^\alpha \)
  \( \alpha \approx 1; A \sim 0. \)

- \( \alpha = \left( \frac{T}{\rho} \right) \cdot \left( \frac{d\rho}{dT} \right) \)
Starting points

1. Start with a charge transfer insulator
   - CT gap: 1.5 eV
2. Doping -> holes -> low-energy spectral weight
3. It’s a superconductor
   - Condensate => Has a $\delta(\omega)$ contribution to $\sigma_1(\omega)$

$$\sigma_1(\omega) = A\delta(\omega) = \frac{\pi \rho_s e^2}{m_e} \delta(\omega)$$

with $\rho_s$ the superfluid density

- London screening (Meissner effect) a consequence of the $\delta(\omega)$ in $\sigma_1(\omega)$
Superfluid density

- Superfluid density, $\rho_s(T)$: fundamental macroscopic quantity of a superconductor.
- Superconducting condensate signaled by spectral weight transfer to $\omega = 0$ delta function.
- Superfluid density, $\rho_s \leftrightarrow$ Strength of the delta function. (Obtained from sum rule [FGT].)
- Superfluid density, $\rho_s \leftrightarrow$ Optical penetration depth. ($\rho_s \sim 1/\lambda_L^2$)

Recall that essentially every conduction electron participates in the $T = 0$ superfluid of a clean metallic superconductor. ($\lambda_L \leftrightarrow c/\omega_p$)
300 K reflectance of LaSrCuO

Photon Energy (eV)

Reflectance

La$_{1.85}$Sr$_{0.15}$CuO$_4$

Frequency (cm$^{-1}$)
Kramers-Kronig of reflectance

- Relates real and imaginary parts of response functions

\[ r = \rho e^{i\phi} = \frac{1 - N}{1 + N} \]
\[ \ln r = \ln \rho + i\phi. \]

\[ \phi(\omega) = -\frac{\omega}{\pi} \text{P} \int_0^\infty d\omega' \frac{\ln[\Re(\omega')/\Re(\omega)]}{\omega'^2 - \omega^2} \]

- Typical data: 30-40,000 cm\(^{-1}\) (4 meV-5 3V)
- Integral: zero to infinity
- \( \therefore \) extrapolations are needed, above and below measured data
- High end gives the most problems
Kramers-Kronig analysis of reflectance: Wooten (1972)

\[ \phi(\omega) = -\frac{\omega}{\pi} \mathcal{P} \int_0^\infty d\omega' \frac{\ln[R(\omega')/R(\omega)]}{\omega'^2 - \omega^2} \]

- How does it do?
KK with power law

La$_{1.85}$Sr$_{0.15}$CuO$_4$

- 0.0
- 0.5
- 1.0
- 1.5
- 2.0
- 3.0
- 4.0

Gao + Tajima

$\sigma_1(\omega) \left( \Omega^{-1} \text{cm}^{-1} \right)$

Frequency (cm$^{-1}$)

Photon Energy (eV)
X-ray optics

- Atomic scattering factors* $f$
- The dielectric function is

$$\epsilon = 1 - \sum_j \frac{4\pi n_j e^2}{m\omega^2} (f_1^j - i f_2^j)$$

- Sum: atoms $j$ at number density $n_j$ and with complex scattering factor $f_j$
- Limiting high-frequency behavior:

$$f_1^j \to Z^j \quad f_2 \to 0 \quad \epsilon \to 1 - \sum_j 4\pi n_j Z^j e^2 / m\omega^2$$

- Reflectance calculated as usual

*See: http://henke.lbl.gov/optical_constants/
LSCO: some vuv data do exist

- How does it do?
IR and CT band independent of bridge
Small variations above 16,000 cm$^{-1}$ (2 eV)

- So far so good.

http://www.phys.ufl.edu/~tanner/ZIPS/datan.zip
La\textsubscript{2-x}Sr\textsubscript{x}CuO\textsubscript{4} \textit{ab-plane} optical conductivity
Bisco 2212 $ab$-plane optical conductivity

![Graph showing the optical conductivity of $(Bi:Pb)_2Sr_2CaCu_2O_8$ as a function of photon energy and frequency at different temperatures.]

- Conductivity ($\Omega^{-1}\text{cm}^{-1}$)
- Photon Energy (eV)
- Frequency ($\text{cm}^{-1}$)

Temperatures:
- 10 K
- 50 K
- 100 K
- 150 K
- 200 K
- 300 K
A set of underdoped crystals

- Area under curves decreases as doping is reduced
- Area smaller below $T_c$
Partial sum rule

• Low-energy carrier density and superfluid density: Partial sum rule

\[ \rho_{\text{eff}}(\omega) \equiv N_{\text{eff}}(\omega) \frac{m}{m^*} = \frac{2mV_{\text{Cu}}}{\pi e^2} \int_0^\omega \sigma_1(\omega') d\omega' \]

• \( e (m) \) free-electron charge (mass), \( m^* \) the effective mass, and \( V_{\text{Cu}} \) the volume allocated to each CuO\(_2\) unit and associated atoms. \((V_{\text{cell}} / Z^*N_{\text{Cu}})\)

Goal: Compare \( \rho_{\text{eff}} \) with \( \rho_s \) for a variety of samples
Two things:

\[ \rho_{\text{eff}}(\omega) = \frac{2mV_{\text{Cu}}}{\pi e^2} \int_0^{\omega} \sigma_{1n}(\omega') d\omega' \]

And

\[ \rho_s(\omega) = \frac{2mV_{\text{Cu}}}{\pi e^2} \int_0^{\omega} \left[ \sigma_{1n}(\omega') - \sigma_{1s}(\omega') \right] d\omega' \]

as \( \omega \rightarrow \omega_{\text{CT}} \)
is linear in $T_c$
is linear in $T_c$
Different $\rho$ scales (1:5)
Superfluid density is small part of total

- \( \rho_s \) increases with \( T_c \). (Uemura plot)
- \( \rho_{\text{eff}} \) increases with \( T_c \). (doping)
- \( \frac{\rho_s}{\rho_{\text{eff}}} \approx 0.2 \)
- In one component picture, the midinfrared absorption is a Holstein sideband, giving

\[
\rho_s = \frac{\rho_{\text{eff}}}{1 + \lambda}
\]

- \((\lambda = \text{mass enhancement factor})\)

\[\Rightarrow \lambda = 4!\]
As a check, use the relation of $\sigma_2$ to $n_s$

- Start with Kramers-Kronig for sigma1 – epsilon1

$$\epsilon_1(\omega) = 1 + 8\mathcal{P} \int_0^\infty d\omega' \frac{A \delta(\omega')}{\omega'^2 - \omega^2}.$$ 

- Do the integral

$$\epsilon_1(\omega) = 1 - \frac{4A}{\omega^2},$$

- Convert to $\sigma_2 = \frac{\omega(1-\epsilon_1)}{4\pi}$

$$\sigma_2(\omega) = \frac{A}{\pi \omega}$$

- As before

$$A = \frac{\omega_{ps}^2}{4} = \pi n_s e^2 / m$$
Compare two quantities:

\[ \rho_s(\omega) = \frac{2mV_{Cu}}{\pi e^2} \int_0^{\omega} [\sigma_{1n}(\omega') - \sigma_{1s}(\omega')] d\omega' \] (Sum Rule)

- which should saturate at high frequencies, and

\[ \rho_s(\omega) = \frac{mV_{Cu}}{e^2} \omega \sigma_{2s}(\omega) \] (London)

- which should be constant at low frequencies.

- And, which should represent the “true” weight of the delta function.
La$_2$CuO$_{4+x}$

$T_c = 40$ K

$\rho_s(\omega)$ vs. Frequency ($\text{cm}^{-1}$)
Superfluid weight comes from low energies

<table>
<thead>
<tr>
<th>Material</th>
<th>From $\omega \sigma_2$</th>
<th>From sum rule</th>
<th>ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Bi:Pb)$_2$Sr$_2$CaCu$_2$O$_8$</td>
<td>0.085</td>
<td>0.080</td>
<td>0.95</td>
</tr>
<tr>
<td>La$<em>2$CuO$</em>{4+\delta}$</td>
<td>0.028</td>
<td>0.028</td>
<td>1.01</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$ (a-axis)</td>
<td>0.101</td>
<td>0.100</td>
<td>0.99</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$ (b-axis)</td>
<td>0.088</td>
<td>0.083</td>
<td>0.95</td>
</tr>
<tr>
<td>I-doped Bi$_2$Sr$_2$CaCu$_2$O$_8$</td>
<td>0.098</td>
<td>0.095</td>
<td>0.97</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$ (transmittance)</td>
<td>0.120</td>
<td>0.122</td>
<td>1.02</td>
</tr>
<tr>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$ a-axis</td>
<td>0.085</td>
<td>0.083</td>
<td>0.98</td>
</tr>
<tr>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$ film</td>
<td>0.062</td>
<td>0.061</td>
<td>0.99</td>
</tr>
<tr>
<td>Y$_{1-x}$Pr$_x$Ba$_2$Cu$<em>3$O$</em>{7-\delta}$</td>
<td>0.057</td>
<td>0.052</td>
<td>0.92</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$CaCu$_2$O$_8$</td>
<td>0.112</td>
<td>0.109</td>
<td>0.97</td>
</tr>
</tbody>
</table>
**Charge transport**

- **Cuprates:**
  - "metallic" dc resistance

  - \( \rho = A + B T^\alpha \)
  - \( \alpha \approx 1; A \sim 0 \).

- \( \alpha = \frac{T}{\rho} \cdot \frac{d\rho}{dT} \)
• \( \rho \sim 150\text{–}300 \, \mu\Omega\text{-cm at 300 K} \)
• Natural to think of a Drude model
• Inadequate once midinfrared absorption kicks in
• Restricts your view to frequencies below about 8 THz (250 cm\(^{-1}\); 30 meV):

• Then, \textit{ab}-plane conductivity is described well by a Drude model.
• The idea of simple free carriers was discarded out long ago.
• Should it be reconsidered in light of experiments showing Fermi Surface reconstruction?
Fermi surface

- Energy dispersion
  \[ E = \frac{\hbar^2 k^2}{2m} \]

- Max $\rightarrow$ Fermi energy
- Fermi surface is a circle in 2D
- Displaced when current flows
Fermi surface reconstruction

- This is what I thought the Fermi surface looked like
Fermi surface reconstruction

• Or maybe this (ARPES results for BiSrCaCuO)
Fermi surface reconstruction

- Or maybe this (ARPES results for BiSrCaCuO)
Fermi surface reconstruction

- Add new zone boundary
Fermi surface reconstruction

- Much smaller area; hole and electron pockets
Fermi surface reconstruction

- Evidence: Shubnikov–de Haas oscillations
Drude conductivity

\[ \sigma(\omega) = \frac{\omega_p^2 \tau}{4\pi(1 - i\omega\tau)} \]

- \( \tau \) - mean free time between collisions.
- \( \omega_p = \sqrt{4\pi ne^2/m} \) - plasma frequency or oscillator strength or spectral weight
- Real part, \( \sigma_{1D}(\omega) \), satisfies sum rule,
  \[ \int_0^\infty d\omega \sigma_1(\omega) = \frac{\pi n e^2}{2m} = \frac{\omega_p^2}{8} \]
- Expect \( n \) constant, \( \tau \) varies with condition (purity, temperature ....)
$\sigma_1(\omega)$ and $\sigma_2(\omega)$ from the Drude model
BSCO 2212 conductivity

Photons Energy (meV)

Conductivity (Ω⁻¹ cm⁻¹)

Frequency (cm⁻¹)

Bi₂Sr₂CaCu₂O₈
Looks like Drude model
Temperature dependence of $1/\tau$ and $\omega_p$

- $1/\tau$: linear in $T$

- Generally

$$\frac{\hbar}{\tau} = 2\pi \lambda k_B T + \frac{\hbar}{\tau_0}$$

- $\lambda = 0.37$
$1/\tau$ is linear in $T$, with $\lambda = 0.35 \pm 0.04$
Most of the Drude weight joins the superfluid
Conclusions

- Only about 20% of doping-induced spectral weight condenses into the superfluid

| Material                  | $\vec{E}||$ | $T_c$ | $\rho_{eff}$ | $\rho_s$ | $\frac{\rho_s}{\rho_{eff}}$ | $\rho_{Drude}$ | $\frac{\rho_s}{\rho_{Drude}}$ |
|---------------------------|------------|------|-------------|---------|-----------------------------|----------------|-----------------------------|
| La$_2$CuO$_{4.12}$        | $ab$       | 40   | 0.15        | 0.028   | 0.19                        | 0.035          | 0.80                        |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$  | $a$        | 85   | 0.40        | 0.10    | 0.25                        | 0.105          | 0.95                        |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$  | $b$        | 85   | 0.44        | 0.090   | 0.20                        | 0.096          | 0.94                        |
| YBa$_2$Cu$_3$O$_7$         | $a$        | 91   | 0.44        | 0.096   | 0.22                        | 0.104          | 0.92                        |
| Tl$_2$Ba$_2$CaCu$_2$O$_8$  | $ab$       | 110  | 0.54        | 0.115   | 0.21                        | 0.13           | 0.88                        |

- Almost all the Drude contribution (assuming the description is correct) condenses
The end
Use of x-ray scattering functions in Kramers-Kronig analysis of reflectance

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Kramers-Kronig analysis is commonly used to estimate the optical properties of new materials. The analysis typically uses data from far infrared through near ultraviolet (say 40–40 000 cm\(^{-1}\) or 5 meV–5 eV) and uses extrapolations outside the measured range. Most high-frequency extrapolations use a power law, \(1/\omega^a\), transitioning to \(1/\omega^4\) at a considerably higher frequency and continuing this free-carrier extension to infinity. The midrange power law is adjusted to match the slope of the data and to give pleasing curves, but the choice of power (usually between 0.5 and 3) is arbitrary. Instead of an arbitrary power law, it is better to use x-ray atomic scattering functions such as those presented by Henke and co-workers. These basically treat the solid as a linear combination of its atomic constituents and, knowing the chemical formula and the density, allow the computation of dielectric function, reflectivity, and other optical functions. The “Henke reflectivity” can be used over photon energies of 10 eV to 34 keV, after which a \(1/\omega^4\) continuation is perfectly fine. The bridge between experimental data and the Henke reflectivity as well as two corrections made to the latter are discussed.

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