Non-equilibrium optical spectroscopy: a new clue to unravel the properties of correlated materials (cuprates)

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i-Lamp
(Interdisciplinary laboratories for advanced materials physics)
• **Time-resolved optical spectroscopy** is a new emerging tool to disentangle the intertwined degrees of freedom

• **Ultrafast coupling with electronic excitations on the fs timescale**
  S. Dal Conte et al., *Science* 335, 1600 (2012)

• **T*(ρ)** line from time-resolved optics
  UNPUBLISHED

• **Superconductivity-induced change of high-energy optical transitions**

• **Real-time probe of phase competition**
People and Collaborations

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

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

• **Equilibrium optical properties of HTSC**
  D. van der Marel (Université de Genève)

• **Samples**
  A. Damascelli (University of British Columbia, Vancouver)
  M. Greven (University of Minnesota & Stanford University)

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

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

• **Time-resolved photoemission** (Duisburg-Essen)
  L. Rettig, U. Bovensiepen
Outline

• Time-resolved optical spectroscopy is a new emerging tool to disentangle the intertwined degrees of freedom

• Ultrafast coupling with electronic excitations on the fs timescale
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• Real-time probe of phase competition
non-equilibrium reflectivity and e-ph coupling

pump probe on metals


optical control

new transient ground state

D. Fausti et al., Science 331, 189 (2011)

coherent excitation

imp. Raman

D. Fausti et al.,
Science 331, 189 (2011)

bandwidth

coherent oscillation

non-equilibrium spectroscopy

recovery of the ground state
Time-resolved optical spectroscopy

Low-fluence (<20 μJ/cm²) and high rep.rate ➞ supercontinuum by a photonic fiber


\[
\frac{\delta R}{R} (\omega, t) = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}
\]

avoid non-thermal destruction of the superconducting phase transition
Equilibrium optical properties of cuprates

extended Drude

$$\epsilon_{D}(\omega) = -\frac{\omega_p^2}{\omega(\omega + M(\omega, T))}$$

scattering rate

$$\text{Im} M(\omega, T) = \hbar/\tau(\omega, T)$$

D.N. Basov et al.  
Rev. Mod. Phys. 85, 471 (2011)
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In metals the frequency-dependence of the electron scattering rate is accounted for by:

\[
\Sigma(\xi) = \int_0^{\infty} d\Omega L(\xi, \Omega; T) \Pi(\Omega)
\]

- In metals e-ph coupling is the only player!

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

- The coupling \( \lambda \) accounts for the critical temperature

\[
\lambda = 2 \int \Pi(\Omega) / \Omega d\Omega
\]
electron-boson coupling in cuprates

In cuprates (optimally and overdoped) the same formalism can be used, provided:

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

Similar features seen in:
- optics
- ARPES
- tunneling

open questions:

- origin of \( \Pi(\Omega) \) and value of \( \lambda \)?
- Is \( \lambda \) enough to account for the critical temperature?
- timescale of e-bos coupling (fs?)

S. Julian *Physics* 5, 17 (2012)
non-equilibrium reflectivity and e-ph coupling

PUMP

Δt

PUMP

PROBE

metal

2-temperature model

\[
\frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e}
\]

\[
\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}}
\]

G/C determines the dynamics in the time domain

non-equilibrium reflectivity and e-ph coupling

pump probe on metals

2-temperature model

\[ \frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e} \]

\[ \frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}} \]

\[ G = \frac{3\gamma_e}{\pi \hbar k_B^2} \int_0^\infty d\Omega \Pi_{lat}(\Omega) [N(\Omega, T_{lat}) - N(\Omega, T_e)] \]

\( G/C \) determines the dynamics in the time domain
non-equilibrium reflectivity and e-ph coupling

2-temperature model

\[
\frac{\partial T_e}{\partial t} = \frac{G(\Pi_{lat}, T_{lat}, T_e)}{\gamma_e T_e} + \frac{p}{\gamma_e T_e}
\]

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\frac{\partial T_{lat}}{\partial t} = -\frac{G(\Pi_{lat}, T_{lat}, T_e)}{C_{lat}}
\]

\[
G = \frac{3\gamma_e}{\pi \hbar k_B^2} \int_0^\infty d\Omega \Pi_{lat}(\Omega) \left[ N(\Omega, T_{lat}) - N(\Omega, T_e) \right]
\]

\[\delta R/R \propto \delta T_e/T_e\]

G/C determines the dynamics in the time domain

in conventional metals: single exponential decay determines \(\lambda\)

Considering non-thermal distribution ($\tau_{ee} > \tau_{e-ph}$):

$$\lambda \langle \omega^2 \rangle = \frac{2\pi}{3} \frac{k_B T_l}{\hbar \tau_{e-ph}}$$

strongly-coupled phonons in cuprates

Time-resolved photoemission

PUMP

photoemitted electrons

cuprate


buckling SCP

breathing SCP

0-300 fs

optimally-doped Bi2212

\[ T_e (K) \]

\[ T_L (K) \]

\[ 700 \]

\[ 600 \]

\[ 500 \]

\[ 400 \]

\[ 300 \]

\[ 200 \]

\[ 0 \]

\[ 1 \]

\[ 2 \]

\[ \text{Delay (ps)} \]

2 TIMESCALES:
• 100 fs (strongly coupled phonons)
• 1 ps (lattice)

are enough 2 timescales to describe the dynamics?

are the electrons decoupled from bosons before 100 fs?
Non-equilibrium optical properties of cuprates

\[
\frac{M(\omega)}{\omega} = \left\{ \int \frac{f(\xi) - f(\xi + \omega)}{\omega + \Sigma^*(\xi) - \Sigma(\xi + \omega)} \, d\xi \right\}^{-1} - 1
\]

\[
\Sigma(\xi) = \int_0^\infty d\Omega L(\xi, \Omega; T) \Pi(\Omega)
\]

\[
L(\xi, \Omega; T_e) + L(\xi, \Omega; T_b)
\]

\[
\frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}
\]

quasi-thermal increase of the scattering time

\[
\delta R/R (10^{-4})
\]

\[
T_e >> T_b
\]

\[
T_e \approx T_b
\]

on which timescale electrons are decoupled from bosons?
electron-boson coupling (T=300K)

\[
\frac{\delta R(\omega, t)}{R} = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)}
\]

optimally-doped Bi$_2$Sr$_2$Ca$_{0.92}$Y$_{0.08}$Cu$_2$O$_{8+\delta}$

double dynamics

simultaneous time and frequency resolution

delay (ps) 0 1 2 3 0.5 1.0 1.5 2.0

energy (eV)

\(\delta R/R\) 0.0000 0.0005

\(\delta R/R\) -0.0005 0.0005
electron-boson coupling (T=300K)

\[ \frac{\delta R}{R}(\omega, t) = \frac{R_{exc}(\omega, t) - R_{eq}(\omega)}{R_{eq}(\omega)} \]

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

simultaneous time and frequency resolution
electron-boson coupling (T=300K)

simultaneous fit in the time and frequency domains

within the time-resolution electrons are never decoupled from bosonic excitations!

how to reproduce simultaneously:
• the two temporal dynamics
• the amplitude of $\delta R/R(\omega)$

$\Pi(\Omega)$ determines both...
electron-boson coupling (T=300K)

S. Dal Conte et al., Science 335, 1600 (2012)

C. Giannetti
Upper bound for the critical temperature

\[ T_c = 0.83 \tilde{\Omega} e^{-\frac{1.04(1+\lambda)}{g(\lambda - \mu^*(1+0.62\lambda)}} \]

\( \tilde{\Omega} \): frequency log-average

\( \mu^* \): effective Coulomb repulsion
\( g=1 \) for s-wave BCS superconductors
\( g<1 \) for d-wave superconductors

assuming \( \mu^*=0, \ g=1 \)

<table>
<thead>
<tr>
<th>bosonic glue</th>
<th>( \lambda )</th>
<th>( \tilde{\Omega} ) (meV)</th>
<th>( T_c ) (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SCP</td>
<td>0.4±0.2</td>
<td>60</td>
<td>2-30</td>
</tr>
<tr>
<td>lattice</td>
<td>0.2±0.2</td>
<td>47</td>
<td>0-12</td>
</tr>
<tr>
<td>electronic</td>
<td>1.1±0.2</td>
<td>87</td>
<td>105-135</td>
</tr>
</tbody>
</table>

the real \( T_c \) is 96 K

\( \sim 70\% \)
short-range AF spin fluctuations?

Ultrafast coupling to high-energy short-range AF spin fluctuations:

• ~2 fs optical scattering rate
• 2-3 fs the inverse of paramagnon width (2-300 meV)

\[ \tau = \frac{\hbar}{\gamma} \quad \hbar = 658 \text{ meV fs} \]
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• Real-time probe of phase competition
The elusive nature of the universal pseudogap

Pseudogap physics $\rightarrow$ interplay between:

- long-range orders
- electronic correlations originating from short range Coulomb repulsion
Long-range orders in the pseudogap

YBCO Nematic
YBCO Stripes
YBCO Magnetic
YBCO Kerr

CDW, NEMATIC, STRIPES, MAGNETIC

Daou 2010
Baek 2012
Fauqué 2006
Xia 2008

Ghiringhelli 2012
Parker 2010
De Almeida 2012
Li 2010
Momentum-selective Mottness

Hubbard Model solved by CDMFT:

- $T^*$ as a crossover between different dynamical regimes, indicating thermodynamic anomalies
- No broken symmetries are necessary
- Momentum selective Mottness in the PG

Rutgers, Columbia, Sherbrooke, Ecole Polytechnique, Sissa
Non-equilibrium spectroscopy

disentangling intertwined degrees of freedom by their dynamics

TR electron-diffraction on 1T-TaS$_2$

Fluctuating CDW in UD La$_{1.9}$Sr$_{0.1}$CuO$_4$

melting of the CDW on the ps timescale


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Interplay between the low- and high-energy scale in HTSC?


spectral weight

$$SW = \int_{0}^{\infty} \frac{\epsilon_{2}(\omega)}{4\pi i} d\omega$$

Ferrel-Glover-Tinkham sum rule

$$SW_{i}^{N} - SW_{i}^{SC} = D - SW_{D}^{N} + SW_{D}^{SC}$$

Kinetic energy in a single conduction band within the nearest-neighbour tight-binding model

$$\langle K \rangle = \frac{4\hbar^{2}V_{Cu}}{\pi^{2}a^{2}e^{2}} [SW_{i}^{N} - SW_{i}^{SC}]$$
Time-resolved optical spectroscopy on Y-Bi2212

\[ \frac{\delta R}{R}(\omega, t) \]

< 10 \mu J/cm^2

Normal state

Pseudogap state

Superconductive state

\[ \text{Bi}_2\text{Sr}_2\text{Ca}_{0.92}\text{Y}_{0.08}\text{Cu}_2\text{O}_{8+\delta} \]

High-energy excitations and superconductivity

Extended Drude model

interband transitions

$\delta R(\omega,t)/R$

Kramers-Kronig constrained fit

$\delta\sigma_1(\omega,t)$

decrease of $\delta\sigma_1(\omega,t)/\sigma_1$ as the doping increases

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F. Cilento et al. in preparation

Superconductivity-induced change of high-energy optical transitions

Real-time probe of phase competition
Competition between pseudogap and superconducting phase

\[ F = \alpha_{SC}|\Psi_{SC}|^2 + \beta_{SC}|\Psi_{SC}|^4 \alpha_{PG}|\Phi_{PG}|^2 + \beta_{PG}|\Phi_{SC}|^4 + W|\Psi_{SC}|^2|\Phi_{PG}|^2 \]

Kinetic eq.:

\[ \frac{d\Phi_{PG}}{dt} = -\lambda \frac{\delta F}{\delta \Phi_{PG}} \]

repulsive \( W > 0 \)

(weak competition)

Conference on:
ULTRAFAST DYNAMICS OF
CORRELATED MATERIALS
14 - 18 October 2013
Miramare, Trieste, Italy
Time-resolved optical spectroscopy is a new emerging tool to disentangle the intertwined degrees of freedom

• Ultrafast coupling with electronic excitations on the fs timescale
S. Dal Conte et al., Science 335, 1600 (2012)

• T* line from time-resolved optics
(anomalous pump-induced decrease of the scattering rate)

• Superconductivity-induced change of high-energy optical transitions

• Real-time probe of phase competition