Ultrafast (but Many-Body) Relaxation in a Low-Density Electron Glass and Spin Systems

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Thorsmolle and Armitage PRL 2010
Helgren, Armitage, Gruner PRB 2004
Helgren, Armitage, Gruner PRL 2002
CUPRATE SUPERCONDUCTORS

Dynamic stabilization?

Enhancing the temperature at which superconductivity is observed is a long-standing objective for materials scientists. Recent tantalizing experiments suggest a possible route for achieving this.

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For 25 years, researchers have tried a myriad of combinations to enhance the superconducting temperature of cuprate superconductors above the record of 134 K set for HgBa₂Ca₂Cu₃O₈ (ref. 1). The hope has always been that just the right chemical variation on the theme of layered CuO planes would drive transition temperatures upwards. The dream is that one may induce superconductivity at or even above room temperature. Writing in *Nature Materials*, Hu et al. report an interesting development in this search. Using intense mid-infrared light pump pulses, they resonantly drive large-amplitude (several per cent of the equilibrium lattice constant) oscillations in the out-of-plane apical oxygens (Fig. 1) in the high-temperature cuprate superconductor YBa₂Cu₃O₆.45. By doing so, they create a transient but highly conducting state at temperatures far above the equilibrium superconducting transition temperature. This state develops a feature in the optical reflectivity called the Josephson plasmon resonance, a key indicator of superconductivity at low temperatures. For YBa₂Cu₃O₆.45, the temperature at which this non-equilibrium effect manifests exceeds room temperature.

The most intriguing interpretation of these observations is that a superconducting, albeit transient, state has been created above room temperature. If true, this would have seminal implications not just for understanding of superconductivity but for understanding of non-equilibrium effects in solids. As one may imagine, however, any report of superconductivity above room temperature is not free of controversy. A few considerations and caveats are therefore in order.

Thermal equilibrium is one of the cornerstones of modern statistical mechanics and condensed-matter physics. By considering and averaging over ensembles that do not evolve in time, microscopic physical phenomena can be related to macroscopic physical laws, in a general way that is independent of the specific model used to describe them. Indeed, the idea of thermal equilibrium even allows us to define concepts as basic as that of temperature itself. But there can be reasons for wanting to push physical systems out of equilibrium and study their behaviour. At least three classes of phenomena and motivations for studying a condensed-matter system out of equilibrium come to mind.

First, experiments are typically performed by ‘pumping’ the system at one time and ‘probing’ it at a later stage. After pumping, one hopes to learn something about the relaxation mechanisms and timescales of the equilibrium phase by watching the system relax and return to equilibrium through the decay of its elementary excitations (scenario 1).

Second, one may drive the system in such a way as to allow access to material configurations (for example, structure or free charge density) that cannot be accessed in equilibrium. Intense pump pulses may therefore change the free-energy landscape and allow a competing phase to be stabilized in a transient fashion (scenario 2). The hope is that the transient phase reflects equilibrium possibilities in a larger generalized parameter space.

Third, one may drive a system to achieve a non-equilibrium phase that cannot exist or is not stabilized without a time-dependent driving field (scenario 3). In this sense, the time-dependent ‘pump’ should be considered a term in the Hamiltonian. Recent work that claims that a light-illumination-driven ‘Flouquet’ topological insulator can be stabilized is of this variety1,4.

The observation reported by Hu et al. is at first sight counterintuitive. They blast a delicate superconducting state with light that dumps energy into the system. One would naively expect to suppress superconductivity, not to enhance it. But the cuprates are complicated materials and it is important to consider the three scenarios described above carefully. The most obvious result of pumping a superconductor with an intense pulse of light is to break Cooper pairs and create a transient conducting state of quasiparticles.

There is a large literature on non-resonant pump–probe studies of the cuprates in which quasiparticle decay is examined in the manner of scenario 1. In the case of the present experiment, the authors put forward reasonable arguments suggesting that they are not exciting quasiparticles in this fashion, but this possibility cannot be ruled out completely. If it is photoexcited quasiparticles, one would still have to explain the high charge mobilities of the order of 1,000 cm² V⁻¹ s⁻¹ observed perpendicular to the CuO₂ planes, where transport is expected to be poor and incoherent in the non-equilibrium regime.

In these experiments, the mid-infrared pump pulse does not illuminate the sample indiscriminately. Instead, it drives a phonon that is known to couple strongly to various kinds of spin and charge order, which generally suppress superconducting correlations6–8. There have been reports of weak superconducting correlations in these compounds to temperatures well above the superconducting transition temperature6. A possibility consistent with scenario 2 is that driving the phonon suppresses

→ Three reasons to go non-equilibrium

1.) Learn about elementary excitations of related equilibrium phase.

2.) Access “phases” not possible with equilibrium material configurations.

3.) New “phases” with no equilibrium analog.

But there is at least one more...
Glasses

Disorder
Interactions
Frustrations

Low Lying metastable states
Slow relaxation
Non-exponential decay
Aging
Memory Effects
Fermi Glass

Disorder $\rightarrow$ Anderson Insulator
Coulomb Glass

Long-range $1/r$ Coulomb interaction: Energy levels are a function of all other occupations
What about the “glass” in electron glasses?

Many metastable quasi-groundstates with large barriers between them...

Hard to reach true ground state; single particle hops eventually will only increase energy...

Multi-particle particle hops are required to lower energy, but these are rare and inefficient

Lots of work on equilibrium properties like hopping conductivity (variable range), tunneling etc. but until recently much less work showing explicitly glassy phenomena (relaxation).
Relaxation in InO$_x$ films (similar in granular metals)

FIG. 1. The dependence of the conductance $G$ on time following a quench from $T=120$ K to $T_m=4.11$ K.

10000 seconds $\sim$ 2.8 hours
What controls the relaxation times?
Relaxation times a function of density?

Connection to orthogonality of quasi-groundstates?
Related to the number of states in a localization volume

Larger at high density.

What about very low densities?
e.g. $10^{18}/\text{cm}^3$

FIG. 3: Typical relaxation time $\tau$, as function of the carrier concentration $n$ for a series of $In_xO$ samples (data are based on the two-dip experiment, see [17] and [28] for fuller details and interpretation). Note the sharp drop of $\tau$ for $n \lesssim 10^{20} \text{ cm}^{-3}$.
Many-body localization

Can a localized system function as its own heat bath; Basko, Aleiner, Altshuler (2005) says “no” (sometimes)

In a strongly interacting many-body system there are no single particle energies, there is only the system energy.

Many-body localization generalizes the idea of mobility edge to the many-body system.
Conclusion

Excite localized charge states with laser, probe with THz

“Ultrafast” relaxation, but slower than natural scales

Powerlaw $t^{-\alpha}$; slow with $|\alpha| < 1$

Many-body effects

Quantum relaxational at low $T$
Lightly doped semiconductors
Lightly doped Si

Si:P @ 39% of $n_c$

$n_p \sim 1.8 \times 10^{18}/\text{cm}^3$

$\xi \sim 13 \text{ nm}$

$n_p^{-1/3} \sim 9 \text{ nm}$

$r_{\text{Bohr}} \sim 2.5 \text{ nm}$

$\varepsilon \sim 14$ ($\varepsilon_{\text{Si}} \sim 11.7$)

Samples previously used for studies of DC hopping conductivity and AC conductivity in phononless regime

Rosenbaum et al. PRL 1983
AC response - “Photon assisted”

Helgren, NPA, Gruner PRB 2004
Less than ~ 7K is phononless regime
AC response

\[ \sigma = A \omega \left[ \frac{\hbar \omega + e^2}{\varepsilon_1 \langle r_\omega \rangle} \right] \]

\[ \omega_c = \frac{e^2}{\varepsilon r_\omega} \]

Helgren, NPA, Gruner PRB 2004
Now PUMP!
Optical Pump - THz Probe

0.8 eV Optical Pump

2.5 meV THz probe

$\Delta T/T \sim -\Delta \sigma/\sigma \sim 10^{-5}$

$\mu J/cm^2 \rightarrow 3.2 \times 10^{15}/cm^3 - 3.2 \times 10^{17}/cm^3$

Pump Fluence  Excited Charge Density

Such experiments probe energy relaxation. Not charge transport
Optical Pump - THz Probe
Trapping times are generally 3-4 ps.
Spectral content of probe pulse peaked at 0.6 THz

No change in transmission phase; $\sigma \to 0 @ \omega \to 0$

Above gap pump is `slow’, but not glassy
Transmission vs. pump/probe delay

5 K

Fluence [μJ/cm²]
- 424.4
- 339.5
- 169.8
- 84.9
- 42.4
- 10.6
- 0.6

ΔT/T [Norm.]

Time [ps]
Power law of transmission decay

\[ \sigma(t) \sim T(t) = A t^{-\alpha} \]

Slow with \(|\alpha| < 1\)

\[ T = 5 \text{ K} \]
Transmission vs. pump/probe delay

\[
\Delta T/T \text{ [Norm.]} \quad \text{Temperature [K]} \n\]

- 5
- 10
- 15
- 20
- 25
- 30
- 35
- 40

\[
\text{Time [ps]} \n\]
Power law of transmission decay

$$\sigma(t) \sim T(t) = A t^{-\alpha}$$

Slow with $|\alpha| < 1$
Power law of transmission decay

\[ \sigma(t) \sim T(t) = A t^{-\alpha} \]
@ Low T and fluence $\Delta T/T$ falls to 30% in 200 ps

\[ \tau_{\text{phonon}} = \frac{1}{\omega_D} e^{(\frac{T_{\text{ES}}}{T})^{1/2}} \approx 9 \text{ picosecond} \]

\[ \tau_M = \frac{\rho_D C}{4\pi \epsilon_1 \epsilon_0} \approx 0.9 \text{ millisecond} \]

phonon assisted

Maxwell time

Relaxation time much longer than naive scale

Totally unrelated to Maxwell time (the time scale for spatial relaxation)
Quantum relaxation regime?

Relaxation finite in limit $T \to 0$ and fluence $\to 0$

No “Hilbert space” localization
Multi-particle relaxation

Rate goes as power > 1;
Evidence for multi-particle relaxation processes

\[ \dot{n} = I_{qp} + 2N\gamma_{pc} - \beta n^2 \]

\[ \dot{N} = I_{ph} + \beta n^2/2 - \gamma_{pc}N - (N - N_{eq})\gamma_{esc} \]
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