Transient cooling of low frequency excitations by impulse perturbations

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Outline

• a simple mechanism to transiently cool down low-frequency excitations by impulse perturbations

• an exactly solvable toy-model that realises such transient cooling
  M.F., Phys. Rev. Lett. 120, 220601 (2018)
  Andrea Nava and M.F., unpublished

• a possible physical realisation: light-enhanced $T_c$ in $K_3C_{60}$
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- Antoine Georges
Rotating wave approximation

- initially, the laser at frequency $\omega$ is off
- when the laser is on, the high-energy mode to be excited requires an energy equal to energy of the mode - $\omega$, as if the mode energy were lower by $\omega$
Rotating wave approximation

the bosonic excitation shifts down in energy by the field frequency $\omega$
with the field on there will be pouring out of low-energy excitations into the high-energy mode.
Rotating wave approximation
at the end of the pulse the population of the low-energy excitations is diminished as if the system were cooler than it was initially.

the missing energy is stored into the high-energy mode.

the system remains trapped into this non-equilibrium state until the high-energy mode decays back.
mutatis mutandis, the same phenomenon might also occur pumping into a Fano-like resonance within a particle-hole continuum. The localised mode has no charge, hence a laser field is just coupled to the p-h continuum.
mutatis mutandis, the same phenomenon might also occur pumping into a Fano-like resonance within a particle-hole continuum.
an exactly solvable toy-model where such selective transient cooling mechanism is realised
Two infinitely connected quantum Ising models coupled to each other

\[ H_{2QIMs} = -\frac{1}{4N} \sum_{i,j=1}^{2N} \sigma_{1i}^x \sigma_{1j}^x - h_1 \sum_{i=1}^{2N} \sigma_{1i}^z \]

\[ -\frac{1}{4N} \sum_{i,j=1}^{2N} \sigma_{2i}^x \sigma_{2j}^x - h_2 \sum_{i=1}^{2N} \sigma_{2i}^z \]

\[ -\Delta \sum_{i} \sigma_{1i}^x \sigma_{2i}^x \]

for each Ising model, any spin is coupled to all others \( \Rightarrow \)

**mean-field is exact in the thermodynamic limit** \( N \rightarrow \infty \)
The $Z_2$ symmetry of the Hamiltonian is spontaneously broken.

$$\langle \sigma^x \rangle \neq 0 \quad \Rightarrow \quad \langle \sigma_2^x \rangle \simeq \frac{\Delta}{h_2} \langle \sigma_1^x \rangle \neq 0$$
for $T \approx T_c$ quite many excitations 1 are thermally populated but very few excitations 2

$n_1$ and $n_2$ are the corresponding populations
excitation 1 will play the role of low-energy modes

excitation 2 will play the role of the high-energy mode

$n_1$ and $n_2$ are the corresponding populations

excitation energies
The “laser pulse”

• in the interval \( t \in [0, \tau] \) we add the time-dependent perturbation

\[
\Delta H(t) = -E_0 \cos \omega t \sum_{i=1}^{2N} \sigma_{1i}^x \sigma_{2i}^x
\]

• \( E_0 \) is the “electric field” amplitude
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- \( \tau \) is the pulse duration

I take \( \tau \) half the Rabi period, with the Rabi frequency \( \Omega = \frac{E_0}{2\pi} \)
The “laser pulse”

• in the interval \( t \in [0, \tau] \) we add the time-dependent perturbation

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\Delta H(t) = -E_0 \cos \omega t \sum_{i=1}^{2N} \sigma_i^x \sigma_{2i}^x
\]

• \( E_0 \) is the “electric field” amplitude
• \( \tau \) is the pulse duration
• \( \omega \) the “laser” frequency

I stick to resonance \( \omega = \epsilon_2(T) - \epsilon_1(T) \)
• during the pulse duration $0 < t < \tau$ the population of mode 2 grows whereas that of mode 1 diminishes

• concurrently the energy of the Ising copy 1 decreases, which is overcompensated by the energy increase of 2

Results at $T = 1.1T_c > T_c$
remarkably, a finite symmetry-breaking order parameter transiently develops despite initially $T > T_c$
Include dissipation via Lindblad’s equations

\[ \omega = \epsilon_2 - \epsilon_1 \]

- I add a dissipative bath at each site, so that mean field remains exact
- six downward jump operators \( L_{n \rightarrow m} = \ket{m}\bra{n} \) with \( n > m = 0, \ldots, 3 \), and coupling strengths \( \gamma_{n \rightarrow m} > 0 \)
- six upward jump operators \( L_{n \leftarrow m} = \ket{n}\bra{m} \) with \( n > m = 0, \ldots, 3 \), and coupling strengths \( \gamma_{n \leftarrow m} \)

\[
\gamma_{n \leftarrow m} = \gamma_{n \rightarrow m} e^{-\beta(E_n - E_m)}
\]
Include dissipation via Lindblad’s equations

\[ |2\rangle \xrightarrow{\omega = \epsilon_2 - \epsilon_1} |3\rangle \xrightarrow{\epsilon_1} |1\rangle \xrightarrow{\epsilon_1} |0\rangle \]

\[ \gamma_{n\rightarrow m} = \gamma_{n\rightarrow m} e^{-\beta (E_n - E_m)} \]

- the dissipative dynamics is controlled by the six parameters \( \gamma_{n\rightarrow m} \)
- if all \( \gamma_{n\rightarrow m} \) are of the same order, the system reaches fast thermal equilibrium, without showing any transient cooling
- if \( \gamma_{n\rightarrow m} \) grow with \( E_n - E_m \), i.e., the high energy excitations decay sufficiently faster than the low energy ones, we do find cooling
Show results for

\[ |2\rangle \rightarrow |0\rangle \]

\[ |3\rangle \rightarrow |1\rangle \]

\[ |2\rangle \rightarrow |3\rangle \]

\[ |3\rangle \rightarrow |2\rangle \]

\[ = 640 \times \]

\[ |2\rangle \rightarrow |1\rangle \]

\[ |1\rangle \rightarrow |0\rangle \]

\[ |2\rangle \rightarrow |3\rangle \]

\[ \gamma_{2\rightarrow 0} = \gamma_{3\rightarrow 0} = \gamma_{3\rightarrow 1} = \gamma_{2\rightarrow 1} = 640 \quad \gamma_{1\rightarrow 0} = 640 \quad \gamma_{3\rightarrow 2} \]

- a pulse profile

\[ E(t) \cos \omega t = \left( \frac{t}{\tau} \right)^2 \exp \left[ 1 - \frac{1}{E_0} \left( \frac{t}{\tau} \right)^2 \right] \cos \omega t \]

- \( E_0 \) = peak amplitude of the pulse
- \( \tau \) = pulse duration
\[ E(t) \cos \omega t = \left( \frac{t}{\tau} \right)^2 \exp \left[ 1 - \frac{1}{E_0} \left( \frac{t}{\tau} \right)^2 \right] \cos \omega t \]

I will show results at \( T = 1.5 \ T_c \) varying \( E_0 \) and \( \tau \) so as to maintain the fluence constant

\[
\text{fluence} \propto \int dt \ E(t)^2 = \text{constant}
\]
\[ \tau = 500 \]
\[ E_0 = 0.19 \]
\[ \sigma = 1 \]

\[ \tau = 1000 \]

\[ E_0 = 0.14 \]

Pulse profile \( E(t) \)
\[ \tau = 1500 \]
\[ E_0 = 0.12 \]

pulse profile \( E(t) \)
\[ \tau = 2500 \]
\[ E_0 = 0.10 \]

pulse profile \( E(t) \)
despite $T = 1.5 T_c > T_c$ the order parameter becomes finite, and remains so for longer times the bigger $\tau$ is at constant fluence
possibly, a physical realisation of that same cooling mechanism

**LETTER**

Possible light-induced superconductivity in K$_3$C$_{60}$ at high temperature

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pump-probe experiment on K$_3$C$_{60}$ molecular superconductor:

- **pump pulse of 300 fs with MIR frequency**
- **optical probe**
Results of the pump-probe experiment

How can we interpret those data?

- equilibrium
- 1ps after the pulse
• the laser pump cleans out thermal excitations from the gap, as if it effectively cooled down the system at low frequency
the transient superconducting-like optical response is observed only when the laser frequency hits a mid-infrared absorption peak
it looks like our cooling mechanism: one pumps into a high-energy mode, and finds that the low-energy excitations are cooled down
we need to find the main actor, i.e., the excitation responsible of the MIR absorption peak
a spin-triplet molecular exciton deriving from the $t_{1u} \text{ LUMO} \rightarrow t_{1g} \text{ LUMO} + 1$ transition strongly pushed down in energy by the positive interplay of Coulomb exchange and Jahn-Teller effect
... specifically, in a molecular calculation

1.1 eV

$\approx 4A_g \approx 4S$

$^2T_{1u} \approx 2P$

Coulomb exchange

$(t_{1u})^2 (t_{1g})^1: 4A_g$

$(t_{1u})^3: ^2T_{1u}$
The absorption process in diagrams

- LUMO
- LUMO+1
- Spin triplet exciton
- S=1 particle-hole pair
- Inter-molecule spin exchange
The absorption process in formulas

\[ \delta \sigma_1(\omega, T) = \gamma \int_0^\infty d\epsilon A_{\text{exc}}(\epsilon, T) \left[ b(\epsilon - \omega, T) \left( \theta(\epsilon - \omega) \chi''(\epsilon - \omega, T) \right) - \theta(\omega - \epsilon) \chi''(\omega - \epsilon, T) \right] - b(\epsilon + \omega, T) \chi''(\epsilon + \omega, T) \]

exciton DOS \quad Bose distribution at temperature T

imaginary part of the dynamical magnetic susceptibility, large since \( K_3C_{60} \) is not far from becoming an AF Mott insulator

not much different from experiment!

note that the width is controlled by \( \chi''(\omega) \) rather than by the exciton lifetime
Assuming Fermi-liquid theory and local equilibrium, the dynamics during the laser pulse is described by the equation of motion

\[
\dot{E}_{qp}(T(t)) = \frac{\partial E_{qp}(T(t))}{\partial T(t)} \dot{T}(t) = c_V(T(t)) \dot{T}(t) \\
= \alpha (\omega - E_{exc}) \delta\sigma_1(\omega, T(t))
\]

Where

\[
\alpha = \frac{\text{fluence}}{\omega \times \text{penetration depth}} \times V_{C_{60}}
\]
• **time-dependent temperature** $T(t)$

\[
\dot{T}(t) = \alpha \frac{\omega - E_{\text{exc}}}{c_V(T(t))} \delta \sigma_1(\omega, T(t))
\]

in such simple modelling the quasiparticle temperature decreases below resonance, $\omega < E_{\text{exc}}$, and increases above
Effective temperature at the end of the pulse:

\[ T_{\text{eff}} = T(t = 300\text{fs}) \]

- \( T_{\text{eff}} \) after the laser shot can be substantially lower than the external value, even lower than \( T_c \).
Conclusions

• a simple and general mechanism to transiently cool down low-frequency excitations might explain the light-enhanced $T_c$ observed in $K_3C_{60}$

• it requires just a high-energy excitation that acts as entropy sink for low-energy ones when the laser is on, and gradually release back that entropy when the laser is turned off
Thank you!